IMPACT OF INCREASE IN NUMBER OF AXIALLY ALIGNED WIRES ON N\textsubscript{2} AND H\textsubscript{2} FLOW OBSTRUCTED BY WIRES ASSEMBLY IN MINIATURIZED CHANNEL

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ABSTRACT

Obstructions placed to the flow have a significant effect on mixing of the fluids, particularly in the miniaturized scales. In this study, the impact of variation in axially aligned wire assembly was investigated for mixing of nitrogen and hydrogen gases towards prospective reactor design for ammonia synthesis. The wires were placed in a square-pitch pattern having 9 (Monolith A), 13 (Monolith B) and 37 wires (Monolith C) wires respectively. For the same inlet velocity of the gases, the contour plots indicate that Monolith B is the optimal design based on the consistent mixing dynamics observed through the velocity contour plot.

Keywords: CFD, Nitrogen, Hydrogen, Mixing, Microchannel

INTRODUCTION

Amongst the important inventions of the 20\textsuperscript{th} century was the discovery of Ammonia (NH\textsubscript{3}). It is used to synthesize fertilizer that increases harvest and saving millions of lives for providing food on the table. Ammonia is commercially produced by the Haber-Bosch process through the reaction of nitrogen (N\textsubscript{2}) and hydrogen (H\textsubscript{2}) gases at a temperature and pressure around 500-650°C and 330 atm, respectively. Low conversion of product yield is usually obtained, while high operating temperature and pressure causes the production process to be costly due to the extreme energy consumption and elevated capital cost of the reactor construction. The high energy poses safety risk of the operation, in addition to greater process control scheme to be utilized for the entire system.

Due to the high market demand, NH\textsubscript{3} production could be improved by utilizing a new route for its synthesis by employing the alteration of their electronic properties subjected to the magnetic induction force. In contrast to the conventional Haber-Bosch process, this method is made possible through conducting the reaction in a microfluidic environment at ambient state. The preliminary synthesis of NH\textsubscript{3} using this proposed method was previously proven at laboratory scale by Yahya et al. [1], in which a microreactor containing Mn-based ferrite catalyst under the magnetic induction zone was able to yield 25% NH\textsubscript{3} at 25°C and 1 atm.

The reaction rate inside the microreactor could be improved by increasing the mixing efficiency which will allow gases to interact more effectively. The enhancement of mixing in the microreactor could have a great influence on the reaction yield and product selectivity [2]. The aim of microfluidic mixing schemes is to enhance the mixing index such that a thorough concentration homogeneity could be achieved.
within a minimum mixing path, hence reducing the characteristic size of microfluidic devices, increasing the contact surface area and time of contact between multiple species [3]. This is done by manipulating the channel geometry or external disturbances that will induce mixing via chaotic advection [4].

Mixing phenomenon can be better understood by studying the flow dynamics of the fluids inside the device. The velocity and temperature of fluids affect mixing; whereas the effect of the bounding wall on mixing length is quite limited [5]. Mixing length reduces as the mixer size decreases, but increases with the increasing inlet pressure as it increases the flow velocity makes the convection effect more pronounced than diffusion [6], [7]. The regular high frequency variation of high vorticity distributions results in effective mixing within shorter length and in shorter span of time [8]. Mixing point is obtained more quickly by decreasing inlet velocity. However, in case of obstructions, flow velocity must be large enough to induce secondary flow and form local turbulence [9], [10].

Reducing the diffusion length between the reactants could enhance mixing inside the microreactor as it is mainly driven by molecular diffusion, where reactor miniaturization leads to low Reynolds numbers in reactor channels. At low Reynolds number, flow may be stratified with streams not interpenetrating each other which results in poor mixing [11]. Using chaotic advection schemes and altering the flow direction laterally can induce stirring and improve mixing even at low Reynolds number. At higher flow rates, the mixing phenomenon is dominated by chaotic advection [12]. The interplay of convective and diffusive transport may be further investigated to enhance mixing keeping in view the constraints of the given length scale. A method to reduce the diffusion length is by splitting reactant fluids into small fluid slices, where the reactant fluids are split into many laminated fluid segments by the geometry of inlet channels into the mixing chamber [13].

Taking the factors mentioned so far in due consideration, the basis of the geometry considered in this work is by creating obstruction for the fluid flow through placing the wires in the axial direction to account for the prevalent gases mixing (of N₂ and H₂). The mixing effectiveness will be investigated via computational fluid dynamics approach (CFD), as it is employed as a design and optimization tool due to its cost-saving, timely, easy for scale-up and safer technique as compared to conducting multiple experimental works [14]. The characterization of the mixing dynamics will provide an insight towards the development of the catalyst placement on the wires that will enhance the reaction rate and increase ammonia yield through the reaction between both gases. In this paper, the prevalence mixing is investigated through the computational fluid dynamic (CFD) approach by analyzing the contour plot of the gas species.

**METHODOLOGY**

A microchannel is to be utilized as a microreaction chamber for the synthesis of NH₃ under the magnetic induction zone at ambient conditions with the presence of a catalyst. The catalyst will be grown onto a wire that serves as its support template, and inserted into the microchannel. As mixing would enhance collisions between H₂ and N₂ that will result in increasing the reaction, the arrangement of the wires in the microchannel is essential to foster the internixing of the gases.

Figure 1 illustrates the dimensional characteristics of the microchannels with the embedded wires. As the axial (YZ-axis) arrangement of the wires resemble a monolith reactor, the term ‘monolithic microchannel’ is therefore used in this study.

The main cylindrical microchannel has a dimension of 10-mm (D) x 50-mm (L) where the gases flow through; while the wire diameter is 0.5 mm each. The wire assembly started at a distance on 3 mm from the inlet and ended 3 mm before the outlet as a mean to induce pre-mixing upon flowing inside. An initial velocity before encountering the obstructions is expected to result in better local turbulence. The types of wires
assembly in the radial direction (XY-axis) are shown in Figure 1, where Monolith A, B and C consist of 9, 13 and 37 wires, respectively. The wires were arranged in a square-pitch manner in the order of loosely-packed to highly-packed configurations.

Steady state simulations were carried out through ANSYS CFX version 15.0, and Table 1 describes the flow properties used in the study. The flow of the gases past each of the monolithic microchannel was set at an inlet velocity of 3.33 m/s to simulate the experimental flow used. The molar ratio of the gases is set at \( \text{N}_2 : \text{H}_2 \) of 1:3. The analysis of the flow dynamics was based upon the velocity and volume fraction contour plots for each gases selected at five different axial locations (YZ-axis), labelled from I to V shown in Figure 1. In this study, the effect of the magnetic induction field and reaction, however, were not considered.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Data</th>
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<tr>
<td>( \text{H}_2 ) mole fraction</td>
<td>0.75</td>
</tr>
<tr>
<td>( \text{N}_2 ) mole fraction</td>
<td>0.25</td>
</tr>
<tr>
<td>Inlet velocity, ( V )</td>
<td>3.33 m/s</td>
</tr>
<tr>
<td>Temperature, ( T )</td>
<td>298 K, isothermal</td>
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<tr>
<td>Pressure, ( P )</td>
<td>1 atm</td>
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<td>Turbulence model</td>
<td>( k-\varepsilon ) model</td>
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Figure 1  Dimensional characteristics of the monolithic microchannels
The mesh sensitivity analysis has been reported in the Figure 2. This has been performed on a geometry of Monolith The value of one of the quantities of interest i.e. average volume fraction of hydrogen at the outlet was observed for the increasing number of mesh elements till near concurrent values were obtained. The CFD model was chosen based on a published study [15] for the channel of comparable size. The simulation results for the CFD models was matched with the experimental data in this study and was in good agreement for the k-ε model.

As depicted by the plot, Monolith A shows an inconsistent mixing dynamics as H₂ velocity fluctuates along the axial direction, despite N₂ appears to be coherent at all locations with a slightly lower speed at the central core. Monolith B, however, provides better mixing since velocities for both gases were well distributed across the channel. Though there were slower pace at the middle region and at the nanowires’ intervals, it was not severe as compared to the latter. Based on this evidence, the catalyst could be proposed to be located on the surface of the wires, with more emphasis to provide much denser placement towards the central core to enhance higher reaction rate for the NH₃ synthesis. Monolith C, on the other hand, displays a wavering velocity range for both gases. Due to the closely-packed assembly of nanowires H₂ moves at a higher speed than N₂, leaving substantially lower speed at the wire intervals. The similarity of the contour hues was only noticeable at location III and IV, which implies to be the only possible location where catalyst could be placed for the NH₃ synthesis to proceed in spite of the shorter mixing length.

**RESULTS AND DISCUSSION**

In elucidating the possible effective mixing dynamics inside the microchannel, the protocol used in the study is by investigating the contour plots through ANSYS Post-Analysis. Two types of contour plots are chosen i.e. velocity of the individual gases and their volume fraction, both in XY- and YZ-axes.

Figure 3 and Figure 4 show the velocity contours of the H₂ and N₂ gases at five axial locations for different types of nanowires assemblies, respectively. A good mixing dynamics was anticipated if the hue of the contour is similar for both gases based on the velocity range shown by the legend on the right side of the figures.
Figure 3  Velocity contours for H₂ flow at various locations in XY-axis
<table>
<thead>
<tr>
<th>Site</th>
<th>Monolith A</th>
<th>Monolith B</th>
<th>Monolith C</th>
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<td><img src="image8" alt="Image" /></td>
<td><img src="image9" alt="Image" /></td>
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<tr>
<td>IV</td>
<td><img src="image10" alt="Image" /></td>
<td><img src="image11" alt="Image" /></td>
<td><img src="image12" alt="Image" /></td>
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**Figure 4** Velocity contours for $N_2$ flow at various locations in XY-axis
Figure 5  Velocity contours for $\text{H}_2$ and $\text{N}_2$ flow at centerline in YZ-axis for Monolith A

Figure 6  Velocity contours for $\text{H}_2$ and $\text{N}_2$ flow at centerline in YZ-axis for Monolith B

Figure 7  Velocity contours for $\text{H}_2$ and $\text{N}_2$ flow at centerline in YZ-axis for Monolith C
In order to visualize from another angle, the contour hues are plotted at point \( Z=0 \) on the \( YZ \)-axis, indicating the radial centreline that the gases flow downstream (Figure-5 and Figure-6). Monoliths A and B show better resemblance in the contours, where the mixture velocities lie in the range of 3.111 to 3.889 m/s, which is similar to the initial set value at the inlet. This translates to the good mixing dynamics as the gases move at the speed where reaction would happen based on the possible effective collision between them to occur. Monolith C, however, shows a different contour trend where the gases at the outer core (nearby the radius of the microchannel) move at faster speed between the velocities of 4.667 to 7.00 m/s, whereas at the central region the gases slow down to a lower than 3.111 m/s. This speed difference can be attributed to the greater effect of wall shear in the central core as compared to the outer shell because the distance between the wall and nearest wire to it was much greater than the distance in between wires [16]. The lesser mixing performance in Monolith C can be attributed to lesser distance between the wires (pitch) on the account of a large number of wires packed inside the microchannel. This has led to the wires behaving as if they are a single-wire assembly with larger diameter, where lesser velocity around its wall are observed with lower segmentation in the fluid flow passing through, thereby resulting in low mixing dynamics.

Taking cue from the velocity contours for mixing, the dynamics for better reaction yield are investigated through the volume fraction contours, as shown in Figure 8 and Figure 9. The similarity of the volume fraction ratio throughout the channel length would predict in better reaction owing to the facts that the gases are available in the requisite molar proportion that would cater for better \( \text{NH}_3 \) synthesis. Should one portion is larger or lesser than the other, this would estimate to lower the yield of \( \text{NH}_3 \) when the actual reaction occurs in the said monolithic microchannel.

In case of Monolith C, the volume fraction for \( \text{N}_2 \) and \( \text{H}_2 \) fluctuates as they flow downstream to a value of around 0.5 in the middle region throughout the channel, thereby not providing the requisite molar proportion for the reactants of 1:3. This difference in proportion of nitrogen and hydrogen gases as compared to the ratio of at inlet can be attributed to their difference in viscosities. With the greater number of obstructions present, the gas with lower viscosity (hydrogen) can develop the velocity profile earlier than nitrogen which allows it to move comparatively faster [17]. The other monolithic microchannels maintain the necessary fraction to a fair extent with Monolith B having an edge towards the former due to the consistencies at each XY-axis sampling slice.

As contour plots in Figure 3 to Figure 9 illustrate the qualitative extent of the mixing scheme inside the various monolithic microchannel configurations, the gradual tone of the contour alone would not explain on how the gas particles move laterally. The degree of swirl or intermingling that proposes mixing dynamics to effectively occur is not well-represented in the changes of hues based on the velocity or volume fraction legend. In order to further confirm whether effective mixing is achieved or not for each assembly, velocity streamlines as shown in Figure-10 are plotted to characterize the dynamics of the gases flowing downstream. Streamlines represent imaginary curves such that the tangent to it at a particular point represents the direction of the instantaneous velocity at that point. It is a path a massless particle would take when placed in that fluid [18]. Hence, the streamline path has been taken to be an indicator of direction of fluid movement in the channel.

The lines of the particle streaks provide the information on the gases particles mobility. If single dots are shown at any spatial distribution across the XY-axis, particle swirl is not anticipated. Whereas, the presence of lines at different length intensities would depicts that \( \text{H}_2 \) and \( \text{N}_2 \) progresses downstream with eddies created, suggesting mixing is enhanced through the pseudo-turbulence phenomenon.
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**Figure 8** Volume fraction contours for H$_2$ flow at various locations in XY-axis
Figure 9 Volume fraction contours for N₂ flow at various locations in XY-axis
Figure 10 Gases component velocity streamlines across the XY-axis
An appreciable degree of swirl is observed at the central core in Monolith A, implying that both H₂ and N₂ is able to generate a possible coalescent chances when they flow across the microchannel. However, 40–50% non-swirl regions existed at outer core away from where the wires are located would infer to the depreciation of the expected NH₃ yield amount in a real reaction system as a result of low particle dynamics. Monolith B, on the other hand, shows better swirl intensities at larger circular region throughout the microchannel compared to the previous assembly. Though near-wall region shows lesser amount of eddy created (10–20% of the total area), the consistency of the swirl observed would anticipate for a better NH₃ yield produced. In contrast to the both monolithic microchannels, Monolith C is somehow deviates from the two where high degree of swirl is noticed at the area where the wires are not fitted in the channel, whereas low intensities occur at the central core. A large discrepancy of velocity values between the two regions predicts that the NH₃ yield to reduce even more than what would be achieved in Monolith A, as the chances of collision between the gases particles do not materialize at the requisite molar proportion of the reactants.

Using the information from this simulation, the possible arrangement of the wires inside the monolithic microchannel would further exemplifies the possibility to enhance NH₃ yield when the actual reaction system is to be developed. By the optimum assembly, which in this case Monolith B, catalyst that would take part in the synthesis of ammonia would be grown on the wires at all its circumference throughout its entire length. The wires will be the catalyst support template, where due to the better eddies created in the optimum configuration, it is anticipated that the NH₃ yield would be similar or better than what is achieved by Yahya et. Al. [1].

CONCLUSION

As inferred from the velocity and volume fraction contours, mixing performance is altered according to the way the wires are arranged. Monolith B is having 13 wires exhibited a better degree of mixing for H₂ and N₂ gases at consistent particle swirl, their velocities and volume fraction of the individual gas at the requisite molar fraction for the better reaction to occur. Monolith A with its nine wires created appreciable mixing intensities, but the low degree of swirl in the outer region away from the wires predicts a lower NH₃ yield to occur. Having too concentrated wires in the central core of the microchannel, Monolith C developed a chance of H₂ gas to be drifting away too fast as compared to N₂, suggesting not only inconsistencies of velocity and volume fraction of the reactants, but also in lower mixing dynamics.

The conclusions were drawn based on the velocity contours and volume fraction contours. These may be regarded as the qualitative results to get an insight into the gaseous mixing inside the microchannel encountered by obstructions. The next area of work would be to quantify the extent of mixing using a mathematical model to get a clearer picture of this mixing process.

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REFERENCES


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