INVESTIGATION OF NANO CATALYST CONFIGURATION IN MICROREACTOR FOR AMMONIA PRODUCTION

By

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CERTIFICATION OF APPROVAL

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A project dissertation submitted to the

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Approved by,

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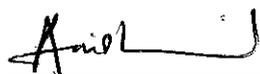
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UNIVERSITI TEKNOLOGI PETRONAS

TRONOH, PERAK

MAY 2012

CERTIFICATION OF ORIGINALITY

This is to certify that I am responsible for the work submitted in this project, that the original work is my own except as specified in the references and acknowledgements, and that the original work contained herein have not been undertaken or done by unspecified sources or persons.

MOHAMAD ZAFRI MAHMUD

ABSTRACT

Ammonia production is continually increasing on every year. The usage of ammonia that is mostly in the fertilizers industry is likely to be the most usage compare to other products of ammonia. Due to that, there are massive research on the improvising the ammonia production. One of the methods is introduction to the microreactor due the low pressure and low pressure operating conditions.

In order to simulate the microreactor configuration, the CFD simulation program is used. CFD simulation is used because the capability to estimate the fluid directions flow based on Navier Stokes equation. It is important since to have a simulation first before fabricate the design because it can save lots of time and money. Besides that, there are a few parameters that need to be determined before jumped to the simulation.

There are three parameters need to be considered in designing the microreactor which are the nanowires configurations, inlet flow direction and phase of the fluid. From the parameters, we can determine the equations and the conditions that need to be consider during the CFD simulation process. Finally, from the simulation, the results then will be compared with the current research to validate and to look for better configuration of microreactor in ammonia production.

ACKNOWLEDGEMENT

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CHAPTER 1

INTRODUCTION

1.1 BACKGROUND OF STUDY

Ammonia production was reported as 131 million metric tons for 2010 (Apodaca, 2011) and estimated to growth about 174 million metric tons in 2015 (PRWeb, 2010). This statement shows that ammonia world demand is high compare to other synthetic gas (syngas) products such as methanol which is 49 million metric tons in year 2010 while acetic acid is 10.7 million metric tons for year 2010 (IHS, 2011). In order to meet the yearly production increment, there are numerous of large scale plants was built and Malaysia is also included. Besides that, there are many researches had been done to increase the production of the ammonia due to shortage of natural gas such as the new way to generate the hydrogen to replace the natural gas using hydrolysis of water other than built a new plant.

The production of ammonia is form by the exothermic reversible chemical reaction of hydrogen and nitrogen (Modak, 2002). Natural gas, hydrogen and steam are the main component in order to produce the syngas which contain the hydrogen and carbon monoxide under the steam reforming reaction. Then, the hydrogen will react with the nitrogen that is extracted from the air to form the ammonia as shown in Figure 1. The process called the Haber–Bosch process which is introducing the iron-based catalyst in early exploration with the operating condition of 150-200 atm of pressure and temperature of 500°C (Modak, 2002). Then, the ruthenium was introduced as a catalyst to lower the energy demand on the operating condition to 100 atm and between 300 and 400 °C (H. Mahapatraa, 1998) besides increases the activation energy and the reaction rate. This is the typical route of the reaction that is use in the large scale plants that exist in the world.

$$\text{Nitrogen} + \text{Hydrogen} \rightleftharpoons \text{Ammonia}$$
$$N_2 \qquad 3H_2 \qquad\qquad 2NH_3$$
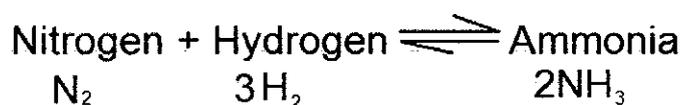
Figure 1 : Ammonia reaction

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Catalyst had been introduced to ensure that the operating condition can be reduced especially on the temperature and pressure. In this 20th century, the development technologies of ammonia synthesis in improving catalyst are growing faster since the demand is increasing every year from the user. In order to develop a new methods and theories, extended studies of the catalyst are important to have more understanding about the characteristics of the catalyst. It has been reported that in ammonia synthesis development, researchers are not only focusing on the component of the catalyst, but there are looking at other detailed areas such the refined characterization techniques, improved kinetic analyses, and new possibilities in theoretical modeling (Claus J.H. Jacobsem, 2000).

1.2 PROBLEM STATEMENT

There are lots of study on the reaction of the hydrogen and nitrogen with the catalyst in a reactor in order to choose the best a way to maximize the conversion rate of the feedstock. Microreactor is one of the reactors that had been introduced to improve the reaction rate of hydrogen and nitrogen to produce ammonia. Microreactor is always related to the micro processing engineering since dealing with the effect of the physical properties such as mass and heat transfer in a microscale chemical technology. Furthermore, the advantages of using microreactor was reported that heat and mass transfer will be more efficient compare to the large scale reactor due to large surface area that exposed to react and it is a continuous reaction (J. Huang, 2009). Moreover, it is also had been proven that microreactor can improve reaction yield and selectivity (Schwalbe T, 2002) and also reaction rate (R.S. Besser, 2003).

In microreactor, it is essential to study about the configuration of the reactor because it will affect how the feedstock will react. In order to determine the best configuration, there are a few parameters that need to be considered such as volume, size and geometry of channels, and also the shape arrangement. There are several studies on the configuration of the microreactor especially on the shape arrangement. It can be conclude that multiple and short packed bed design will increase the surface area for the catalyst to the volume of feedstock (Ratchananusorn, Chemical Engineering and Processing: Process Intensification, 2011) while minimizing the pressure drop (J.C.

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Ganley, 2004). In this project report, it is important to know the effect of microreactor configuration on the chance of molecules to reside on the catalyst surface.

1.3   OBJECTIVE

The main objectives of this project are:

1.   To computationally design the configuration of nanowire catalyst microreactor.

2.   To investigate the effect of nanowire catalyst configurations on the distribution of reactant gas.

1.4   SCOPE OF STUDY

Using computational fluid dynamic (CFD) modeling, the effect of the geometric structure towards the reaction of the feedstock can be reviewed. Hence, from the results, the most suitable configuration for hydrogen to react with nitrogen to form ammonia can be predicted. It is necessary to carry out CFD simulation for the micro reactor so that the assumption can be verify and correct. The CFD simulation for this project will be focusing on the geometry of the channels in the microreactor towards catalytic reaction. Different type of geometry should give the different type of flow distribution and different surface area that will expose to the certain volume.

Besides that, in this project, the modeling will be in 3-Dimensional (3D) geometrical model. The project will be focusing on how the characteristic of the flow after going through certain shape in a small scale system. The geometric that is proposed in this project was the straight line, square and triangle shape inside the channels. Then, from the result, we can see at which area of the geometric shapes will occur the most reactions of the feedstock.

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CHAPTER 2

LITERATURE REVIEW

In this project, the main focus only on the relationship between the microreactor configuration and the effect of reaction site for ammonia production. The feedstock will be nitrogen and hydrogen to be the main component to study about this 3D simulation of the geometry effects as the reaction is occurred in two of the three most prevalent methods to produce ammonia which are Haber-Bosch and indirect electrochemical dissociation (Sahu, 2010). This simulation will focus on the flow distribution for each geometric shape and will come out with the best result to react the gas and gas phase with the catalyst as the conclusion.

2.1   GEOMETRY SHAPES SELECTION

There are few parameters that need to take for consideration in this project. It is important to start with the geometry shapes selection in the channels before designing microreactor. There are few shapes that had been experimented with different functions and one of them is cone with shaped flat bottom. In catalyst trap reactor, the trapezoidal arrangement of four posts cone with shaped flat bottom is created in order to trap the catalyst with approximately 30 catalyst particles in each trap. From the results, it can be concluded that catalyst trap microreactor can be determined by the arrangement of geometric structured to capture the catalyst (Besser, 2009).
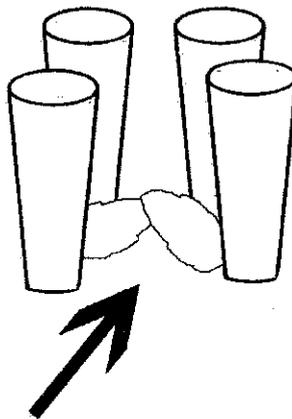
Figure 2 : Catalyst trap reactor geometry configuration

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2.2   INLET FLOW DIRECTION

. The other parameter influencing the design is the direction of inlet flow into microreactor. There are two types of flow direction which are vertical and horizontal. In the horizontal arrangement, there are two direction of flow on microreactor which are the axial flow (horizontal with parallel to the long axis of the tube) and the cross-flow (horizontal with perpendicular to the long axis of the tube). Both types can be use for heterogeneous catalyst testing and to determine the kinetic parameters (Ajmera, 2001). Then, for the conclusion, microreactor with the axial and cross flow both of the arrangement can determine the catalyst testing trap and the kinetic parameters.
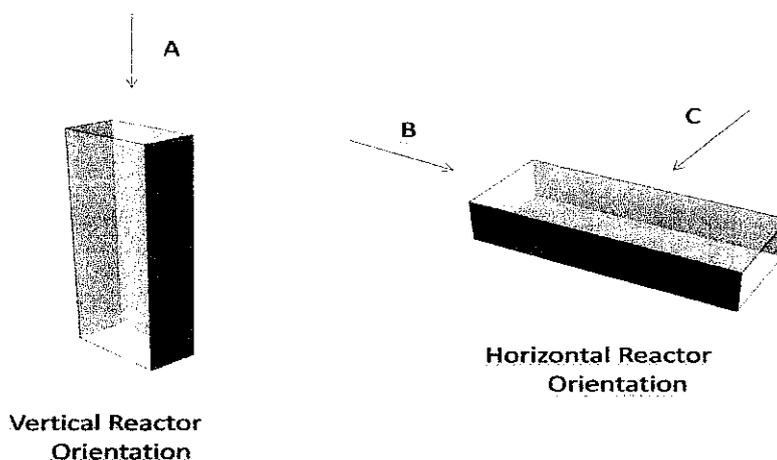
Figure 3 : Different microreactor inlet flow direction. A) Vertical, B) Horizontal and parallel, C) Horizontal and perpendicular

2.3   PHASES OF INLET

Besides that, for each phases of inlet will give different results of the flow distribution and also the mixing behavior. For example, in gas and liquid phases, the best geometry structured will be the one with triangle shape compare to square (Warin Ratchananusorn, 2011). This is because the triangular shape gave slightly better effects on the hydrodynamic behavior and as the result, mass transfer will be faster compare to

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the square ones. Due to that, the phases for the inlet need to be determine first since it might give the different effects with the different geometric shapes.
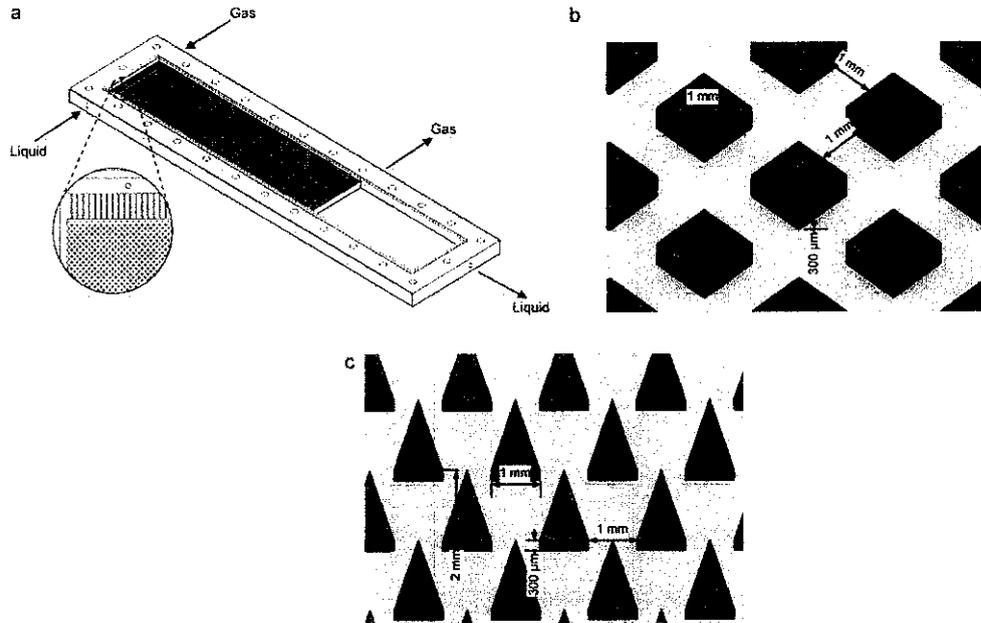
Figure 4 : Liquid-Gas inlet phases of microreactor and the geometry shape

2.4 NAVIER STOKES EQUATIONS

Based on the study, it can be conclude that there are 3 parameters that will be considered in this simulation. There are the geometry structure, the flow direction and lastly the phases. Then, before the modeling can be done, it is important to know the equations that will be used to determine the flow distribution. First of all, using the CFD simulation, it is important to understand the **Navier Stokes equation** because it is the fundamental basis of the CFD problems. Navier Stoke's equation is important because in simulation, it can define any single phase fluid flow.

The derivation of the Navier–Stokes equations begins with an application to the conservation of momentum, mass and energy equations and written as the portion of the fluid behavior solutions (Batchelor, 1967). The general form of the equations of fluid motion is based on the conservation theory can be shown in Figure 5.

$$\rho \left(\frac{\partial \mathbf{v}}{\partial t} + \mathbf{v} \cdot \nabla \mathbf{v} \right) = -\nabla p + \nabla \cdot \mathbb{T} + \mathbf{f}.$$

Figure 5 : Navier Stokes equation (general)

Where,

- $\mathbf{v}$ is the flow velocity,
- ρ is the fluid density,
- p is the pressure,
- $\mathbb{T}$ is the (deviatoric) stress tensor ,
- $\mathbf{f}$ represents body forces (per unit volume) acting on the fluid ,
- ∇ is the del operator.

From the equation, acceleration and composed of time dependent or convective effects (also the effects of non-inertial coordinates) are describe at the left side of the equation while on the right side of the equation is in effect a summation of body forces (such as gravity) and divergence of stress (pressure and shear stress). Navier Stokes general equation then can be used to calculate the fluid motion in any direction. As for example, the general equation is also can be applied to any non-relativistic continuum which is known as Cauchy momentum equation (Nadish Saini, 2012).

2.5   EULER EQUATIONS

In **Euler equations**, it is a combination of a set of equations governing inviscid flow in fluid dynamics. Again, the equations represent conservation of mass (continuity), momentum, and energy and the equations are related to the Navier Stokes equations based on the viscosity and heat conduction. Although Euler equations only derived the mass and momentum equations, the fluid dynamics concept of study is always refers to the conservation of mass, momentum and energy. From the three combinations of conservation, then it become "the Euler equations" (Anderson, 1995).

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Euler equations are always written in two main forms which are the "conservation form" and the "non-conservation form". While the volume is fixed in space, the conservation form will be used and it will focus on the physical interpretation of the equations. For the non-conservation form, the equations will emphasizes on the changes to the state of a control volume as it moves with the fluid.

The Euler equations are mostly applied to compressible as well as to incompressible flow. This is can be done by using the appropriate equation of state or assuming that the flow velocity divergence field is zero, respectively.

In differential form, the equations can be shown in the Figure 7.

$$\frac{\partial \rho}{\partial t} + \nabla \cdot (\rho \mathbf{u}) = 0$$

$$\frac{\partial \rho \mathbf{u}}{\partial t} + \nabla \cdot (\mathbf{u} \otimes (\rho \mathbf{u})) + \nabla p = 0$$

$$\frac{\partial E}{\partial t} + \nabla \cdot (\mathbf{u}(E + p)) = 0.$$

Figure 6 : Euler's equation

Where,

- ρ is the fluid mass density,
- u is the fluid velocity vector, with components u, v, and w,
- $E = \rho\, e + \frac{1}{2} \rho\, (u^2 + v^2 + w^2)$ is the total energy per unit volume, with e being the internal energy per unit mass for the fluid,
- p is the pressure,
- $\otimes$ denotes the tensor product, and
- 0 being the zero vector.

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CHAPTER 3

METHODOLOGY

Before entering the simulation part, there are three parameters that need to be determined first. First of all, it is important to determine the nanowires configuration in the microchannel. By using CFD simulation, there are two types of nanowires configuration in the flow channels that will use as the project studies which are Type A and Type B. Secondly, it will consider only the horizontal flow direction for the inlet stream to the reactor which means that the flow might be from the left to the right side or vice versa with parallel to the long axis of the tube. Finally, the phases of the inlet stream to the microchannel also need to be determined and for this modeling, hydrogen and nitrogen will be the inlets which are both in the gas phases. The methodology of the simulation is shown in Figure 2.

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┌─────────────────────────────────────────────────────┐
│                    1. Start                          │
└─────────────────────────────────────────────────────┘

┌─────────────────────────────────────────────────────┐
│        2. Study and Identify the project background  │
└─────────────────────────────────────────────────────┘

┌─────────────────────────────────────────────────────┐
│   3. Model Development according to the selected parameters │
└─────────────────────────────────────────────────────┘

┌─────────────────────────────────────────────────────┐
│              4. Simulation using CFD model           │
└─────────────────────────────────────────────────────┘

┌─────────────────────────────────────────────────────┐
│   5. Validation by compare the results with previous study │
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│                 6. Implementation                    │
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┌─────────────────────────────────────────────────────┐
│                    7. End                            │
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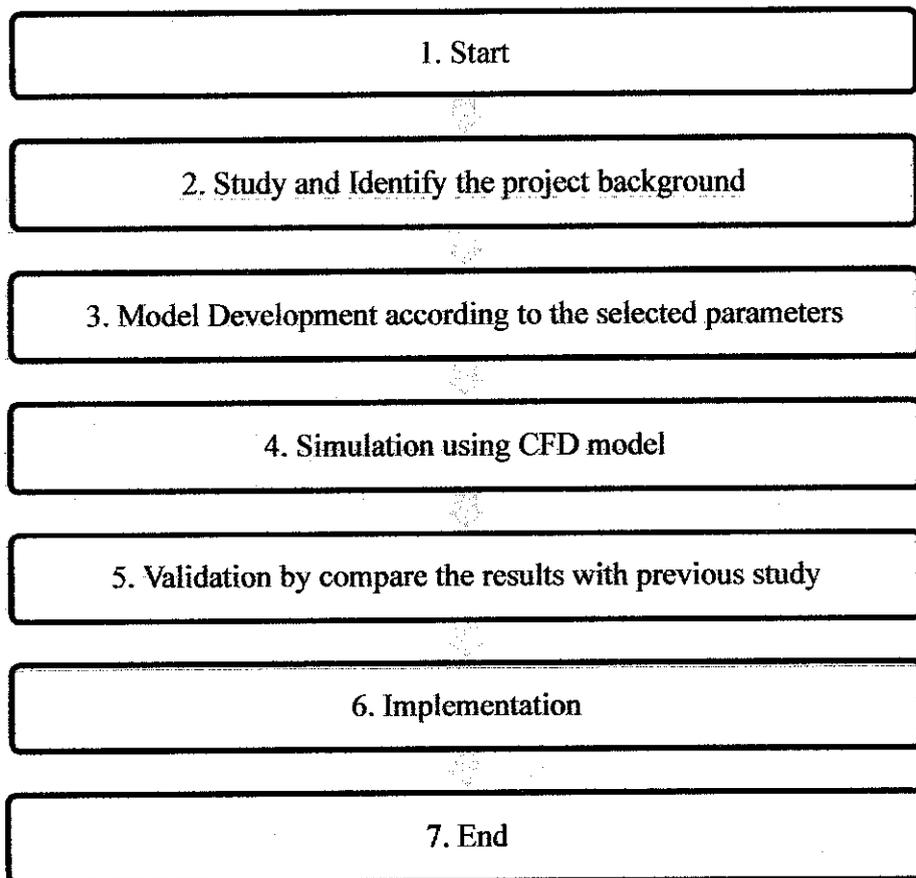
Figure 7 : Research Methodology Diagram

3.1   COMPUTATIONAL FLUID DYNAMIC (CFD)

Computational fluid dynamic (CFD) simulation is important in order to have the real situation of the fluid motion before pattern the real design. As going through the simulation, there must be a few steps to be done in order to get the final results. Besides that, the steps must be followed accordingly before move to the other steps. So, to have a deep understanding on the CFD simulation before using it is essential.

The steps involve in the CFD simulations can be divided into 3 major steps which are pre processing, solver and post processing. Pre processing steps is where the goals and the problems have to be determined. Then, follow by the solver which is in this step, it is important to have the equations and the mathematical manipulation in order to calculate flow. Lastly, the post processing process is the step that will solve all the equations based on the goals and problems and come out with the results.

The flow chart of the CFD simulation steps can be shown in the Figure 9.

PRE PROCESSING		
Simulation Goals		Input of Flow Problem

SOLVER		
Flow Equation	Mathematical Manipulation	Algebraic Equations

POST PROCESSING			
Geometry & Grid Display	Vector Plots	Line & Shaded Contour Plots	2D and 3D Surface Plots
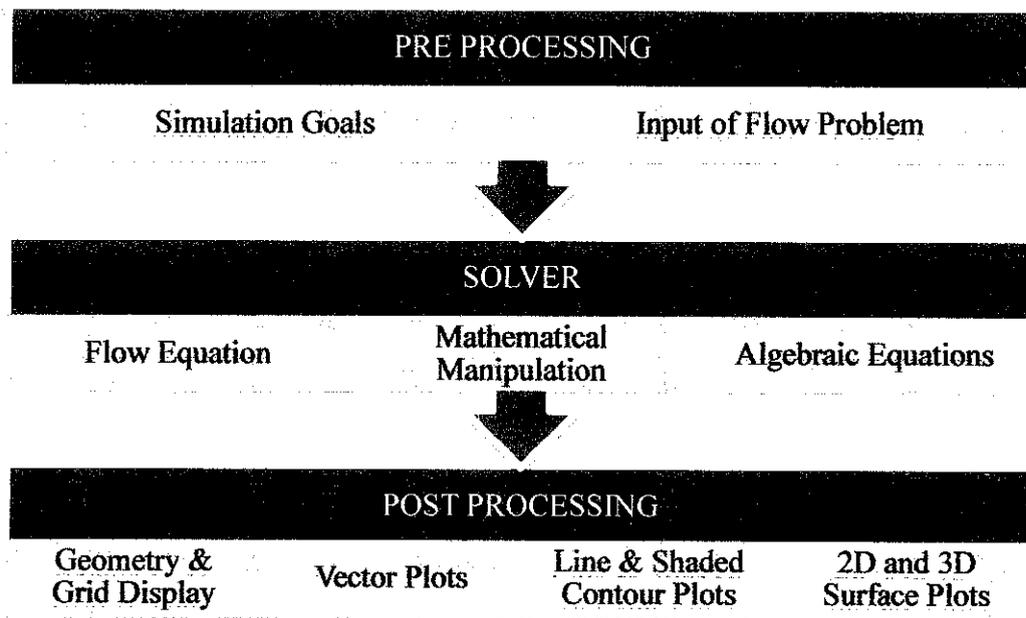
Figure 8 : CFD simulation steps

3.1.1 MODEL GEOMETRY

Before undergo the pre processing step, it is important to determine and develop the geometry in order to simulate the two phase flow problem. For this simulation, the geometry had been developed using Design Modular (DM) since DM is one of the medium to design the 2D and 3D model. 3D model had been chose to develop the microchannel geometry because the tubular effect inside a cylinder can be simulated more precise compare to 2D model.

3.1.1.1 Specifications

All geometry specification for microchannel were already given by OneBaja project, while other specifications had to be made based on assumptions related to the capabilities of ANSYS 14.0. OneBaja specified a packed bed reactor in micro scale called the microreactor. This model geometry consisted of a tube channel called microchannel laid horizontally and packed with nanowires catalyst. The microreactor dimension reported were a length of 2.5 cm while the microchannel diameter inside the microreactor will be 10 μm.

Since the microreactor description from OneBaja project lacked details for the specific configuration placement of nanowires inside the microchannel, a series of configuration models were chosen to test. These models were designed in Design Modular (DM) based on the dimensions specified by the project. Verification of the orientation of nanowires was verified according to the suitability of the configuration towards the microchannel.

3.1.1.2 Dimensions

In DM, the 3D model of microchannel was developed only using the symmetry model which is only half of the microchannel instead of full cylinder to reduce the simulation time. Besides that, the length of microchannel was only 2 μm from the inlet and the diameter of microchannel remained the same as 10 μm. Then, for the nanowires geometry, it is reported that the nanowires have a 15.0 nm of diameter and minimum of 2.0 μm height of nanowires inside the microchannel. Additionally, no report was made

to address the nanowires configuration and the distances between the nanowires inside the microchannel.

Figure 9 shows the 3D modeling of microchannel that had been developed using DM.
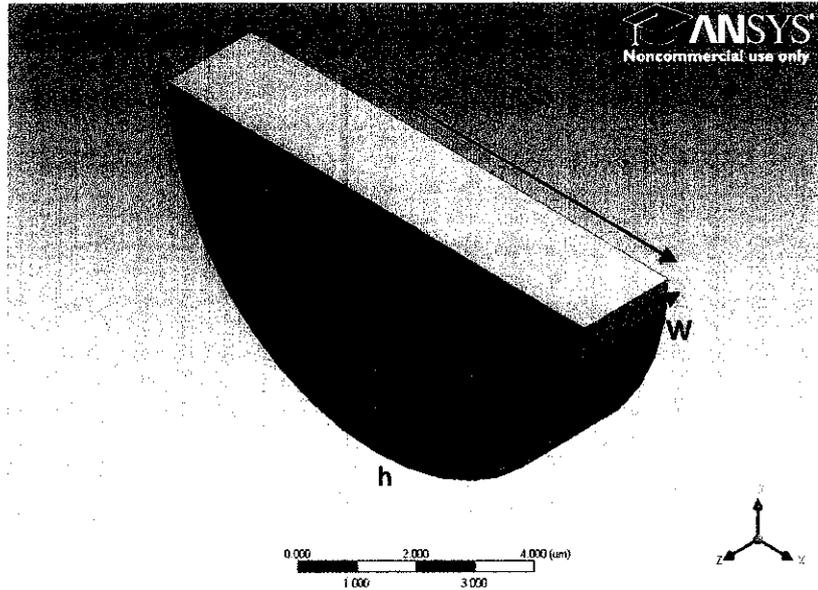
Figure 9 : Microchannel dimensions

Dimension	
d	10 µm
W	2 µm
h	15 ηm

3.1.1.3 Configurations

There are two types of nanowires configuration that were proposed for the simulation which are Type A and Type B. The first model, Type A can be described as the parallel nanowires orientation which is place side by side along the z-axis with 1 µm distance between the nanowires inside the microchannel surface. The nanowires were placed with 45° to the center along the inner surface. This model demonstrated a small gap between the two nanowires along the z-axis while leaving a big gap for the x-axis. Finally, from the details of the dimension for nanowires, total of 6 nanowires were placed along the 2 µm length of microchannel, for the first model.

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Figure 10 shows the design for the nanowires configuration inside the microchannel that had been developed.
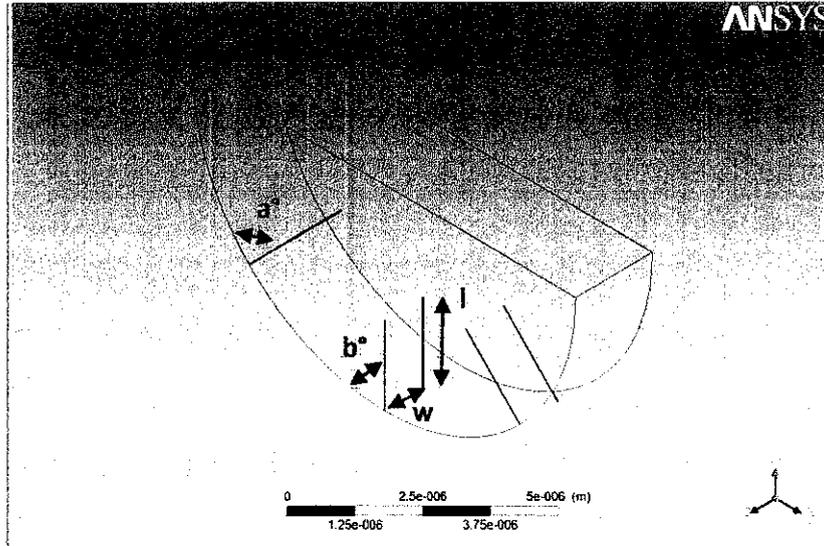
Figure 10 : Nanowires configuration for Type A

Dimension	
l	2 μm
w	1 μm
a°	45°
b°	90°

The second model, Type B can be described as diagonally nanowires orientation, in which the nanowires were placed along the inner surface of microchannel in a zig-zag pattern to leave open space between the nanowires. The location of the nanowires inside the microchannel was also determined by the degree of the nanowires towards centre which is the first row will be 22.5° and the following row is 45°. The orientation was repeated as the first two rows along the microchannel. Lastly, the total of 5 nanowires had been placed inside the microchannel for the second model.

Figure 10 shows the design for the nanowires configuration inside the microchannel that had been developed.
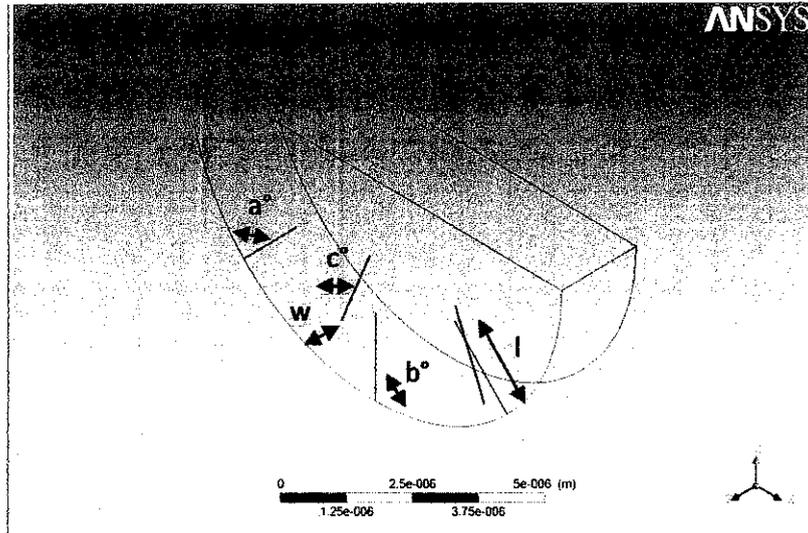
Figure 11: Nanowires configuration for Type B

Dimension	
l	1 μm
w	1 μm
a°	45°
b°	90°
c°	22.5°

3.1.1.4   Velocity

Based on OneBaja project, there are 3 different inlet velocities for both nitrogen and hydrogen inside the microchannel that need to be studied. From the simulation results, the volume fraction and the velocity profile will be analyzed to give the best nanowires configuration for better ammonia production. Calculation shows the conversion of volumetric flowrate, Q to velocity, v for 3.33 ml/min given by the OneBaja project.

Conversion of Volumetric Flowrate, Q to Velocity, v :

$$Volumetric\ Flowrate, Q = \frac{volume}{time} = \frac{3.33\ ml}{min} \times \frac{1\ min}{60\ sec} \times \frac{1\ lit}{1000\ ml} \times \frac{1\ m^3}{1000\ lit} = \frac{5.55 \times 10^{-8}\ m^3}{sec}$$

$$Area, A = \pi j^2 = \pi(5 \times 10^{-6}m)^2 = 7.854 \times 10^{-4} m^2$$

$$Velocity, v = \frac{Q}{A} = \frac{5.55 \times 10^{-8}\ m^3/sec}{7.854 \times 10^{-6}m^2} = 706.65\ \frac{m}{s}$$

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Table 1, shows the inlet velocity for nitrogen and hydrogen inside the microchannel for the three different velocities based on the calculation.

	Volumetric Flowrate, Q	Velocity, v
	1.11 ml/min	235.50 m/s
Velocity	2.22 ml/min	471.10 m/s
	3.33 ml/min	706.65 m/s

Table 1 : Inlet velocity for nitrogen and hydrogen

3.1.2 MESHING MODEL

In pre processing step, or also called meshing where the area of study must be determine in this step before going through the other steps. ANSYS 14.0 simulation will be calculated and come out with the meshing model for the specific area of study. It is important because after determined the area of study such as the study of fluid motions on the specific wall or fluid motions at the nozzles, the condition of the geometry for each faces will be setup before going through the solver step. ANSYS FLUENT that had been provided for the simulation is an academic version and there are few limitations that need to be followed in the meshing setup before going to the FLUENT setup.

```
       For full Legal Notice, see documentation.

Build Time: Oct 25 2011 13:08:50  Build Id: 10398
Loading "C:\PROGRA~1\ANSYSI~1\v140\fluent\fluent14.0.0\lib\fl_s1119-64.dmp"
Done.


       ----------------------------------------------------------
       This is an academic version of ANSYS FLUENT. Usage of this product
       license is limited to the terms and conditions specified in your ANSYS
       license form, additional terms section.       -----------------------------------------------------


Cleanup script file is C:\\Documents and Settings\\Guest\\cleanup-fluent-WS-03-4960.bat


> Reading "\"| gunzip -c \"C:\4. Eulerian Model - Parallel (471ms)\Eulerian Parallel (471.10ms
  491900 tetrahedral cells, zone  3, binary.
    6377 triangular wall faces, zone  1, binary.
  975488 triangular interior faces, zone  2, binary.
```

Figure 12 : Software version

3.1.2.1   Mesh parameters

At this stage, it is important to determine the condition of the geometry such as the inlet, outlet and the wall. This is important to ensure that it can be understood by the solver that there are certain conditions that need to be considered in solving the equation in solver stage. For the microchannel, since the fluid is flowing through z-axis, the inlet and outlet faces had been specified at the front and back of the geometry. The symmetry faces then being specified at the top of the geometry since the geometry is only drawn half of the microchannel. Lastly, the outer side of the channel was defined as the wall face. Figure 12 shows the condition of faces that had been determined.
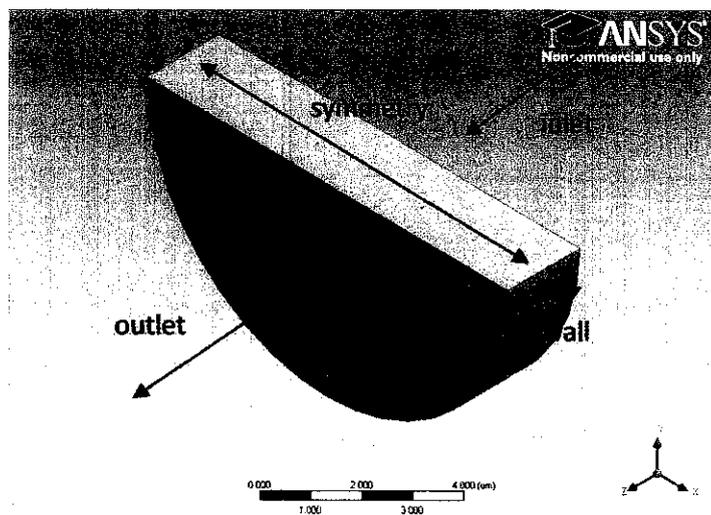
Figure 13 : Geometry condition

Next, after determined all the conditions for the geometry inlet, outlet, wall and symmetry, model mesh was generated. The details of mesh then were being analyzed and modified to give the suitable setup according to the limitation of ANSYS 14.0 for the model before entering the next stage.

The standard mesh parameters can be separated into seven categories which are default, sizing, inflation, assembly meshing, patch conforming options, advanced options, and defeaturing. Due to the nature of the geometry, assembly meshing, patch conforming options and defeaturing were not changed from the default settings which included no assembly meshing, program controlled for triangle surface mesher in patch

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conforming options and automatic mesh-based defeaturing. Similarly, there was no need to explore advanced options, so this category remained untouched.

There are 3 setting in the defaults category which is Physics Preference, Solver Preference, and Relevance. The physics preference was chosen to be CFD (Computational Fluid Dynamics) due to the project goal specification. Then, in the solver preference, FLUENT was chosen for reasons of using CFD as the physics preference at the beginning. ANSYS is a multiphysics simulation program that can be applied for many types of problems or systems, and it has several solver approaches to address the solutions for these different types of problems and systems. In CFD preference, there are two types of solutions that are that is given by defaults from ANSYS 14.0 which are CFX and FLUENT. Both of these models can be used to solve the fluid flow problems and for this project, FLUENT CFD had been chosen.

The fineness of the mesh and other additional quantitative conditions were specified in the relevance qualitatively option. Due to some limitations in the software license, the elements generated cannot exceed 512000 and the setup needs to meet the limitations before entering the next stage. In ANSYS meshing software, the relevance option defines a scale from -100 to 100, with zero being the "moderate" mesh size. As the results, the relevance scale was set at -99 for Type A and -100 for Type B as the base point.

Then, sizing category was maintained as the given default conditions. The only main difference was specifying the relevance center option as coarse. There are 3 types of relevance center options which are fine, medium and coarse. In order to increase the uniformity overall, the relevance center need specified as fine. This is important to setup the best mesh uniformity before stepping to the next stage because more accurate results can be calculated for the meshes with high uniformity but due to limitation, the relevance center is set to be coarse.

Figure 13 shows the details of mesh for Type A and Type B with changed the relevance center to "fine".

Type A

Type B

Details of "Mesh"			Details of "Mesh"	
Defaults			**Defaults**	
Physics Preference	CFD		Physics Preference	CFD
Solver Preference	Fluent		Solver Preference	Fluent
Relevance	-100		Relevance	-99
Sizing			**Sizing**	
Use Advanced Size Function	On: Curvature		Use Advanced Size Function	On: Curvature
Relevance Center	Coarse		Relevance Center	Coarse
Initial Size Seed	Active Assembly		Initial Size Seed	Active Assembly
Smoothing	Medium		Smoothing	Medium
Transition	Slow		Transition	Slow
Span Angle Center	Fine		Span Angle Center	Fine
Curvature Normal Angle	Default (36.0 °)		Curvature Normal Angle	Default (35.7610 °)
Min Size	Default (1.4197e-008 m)		Min Size	Default (1.4082e-008 m)
Max Face Size	Default (1.4197e-006 m)		Max Face Size	Default (1.4082e-006 m)
Max Size	Default (2.8395e-006 m)		Max Size	Default (2.8164e-006 m)
Growth Rate	Default (1.20)		Growth Rate	Default (1.20050)
Minimum Edge Length	1.5e-008 m		Minimum Edge Length	1.5e-008 m
+ Inflation			**+ Inflation**	
Assembly Meshing			**Assembly Meshing**	
Method	None		Method	None
Patch Conforming Options			**Patch Conforming Options**	
Triangle Surface Mesher	Program Controlled		Triangle Surface Mesher	Program Controlled
+ Advanced			**+ Advanced**	
+ Defeaturing			**+ Defeaturing**	

Figure 14 : Details of mesh with "fine" relevance center setup

3.1.2.2   Mesh statistics

Based on the details of mesh, quality of meshing can be studied and improved for better results. In the mesh statistics category, there are two main options which are nodes and elements. As the results from the generated mesh, Type A showed that the nodes that being used for the mesh is 67130 and 377988 for the elements while for Type B, the nodes and elements is 86608 and 491900. Then, in statistics category, mesh metric options was changed to orthogonal quality to check the mesh quality for the geometry. The minimum, maximum, average and the standard deviation line for the element metrics was showed to see the results of the quality.

Figure 14 shows the statistics category details for Type A and Type B.

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Type A | Type B

- Statistics			- Statistics	
Nodes	86608		Nodes	67130
Elements	491900		Elements	377988
Mesh Metric	Orthogonal Quality		Mesh Metric	Orthogonal Quality
Min	0.188379079949749		Min	0.228463971759062
Max	0.995454715021224		Max	0.996289637247543
Average	0.854680352420573		Average	0.853192275590833
Standard Deviation	0.088432928267112		Standard Deviation	8.96803613917943E-02

Figure 15 : Mesh statistics results

Orthogonal quality is the mesh statistics to show that the better quality for the generated mesh is when the element metric value is near to 1. Then, from the graph, the results showed that the quality of the mesh was near to 1 since the most of the elements number are more than 0.5 of element metrics. For Type A, the maximum number of element metric is 0.995 and the minimum is 0.188. The highest number of elements is 199247 out of 491900 are when the element metrics is at 0.88 based on the results from the graph. Then, for type B, the maximum number of element metric is 0.996 and the minimum is 0.228 while the peak number of elements is 148806 out of 377988 is when the element metrics is at 0.88.

Graph for the number of elements versus the elements metric for Type A and Type B is shown in Figure 15.

Type A

Mesh Metrics

Controls

Tet4

Type B

Controls
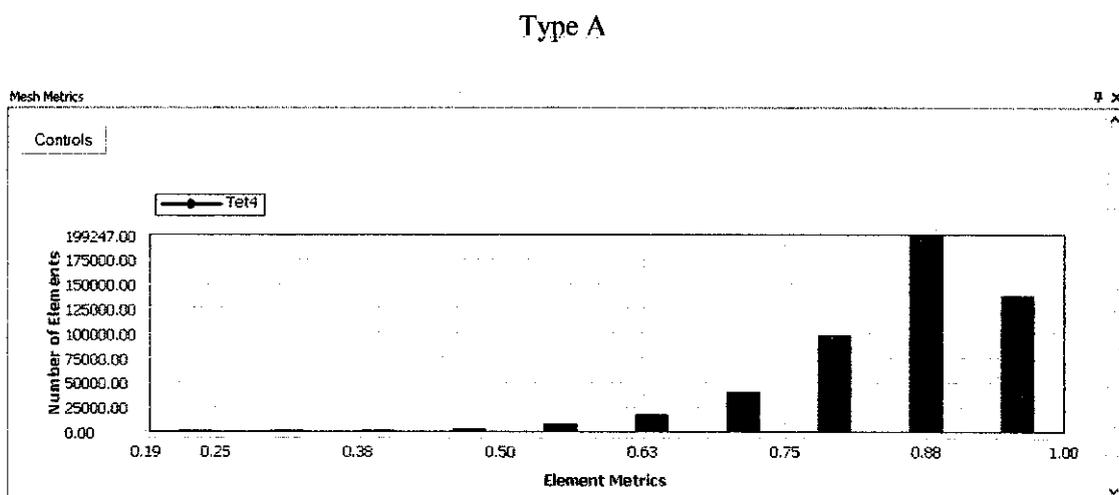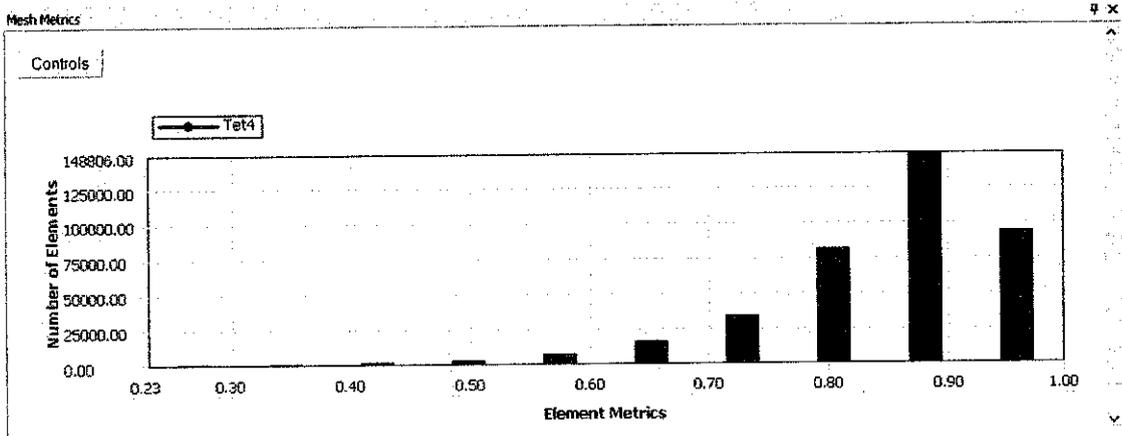
Figure 16 : Graph of Number of elements vs. Element metrics for Type A and Type B

3.1.2.3   Mesh generated model

Type A generated mesh geometry for inlet, outlet and isometric view are showed in Figure 16.

Inlet view

20
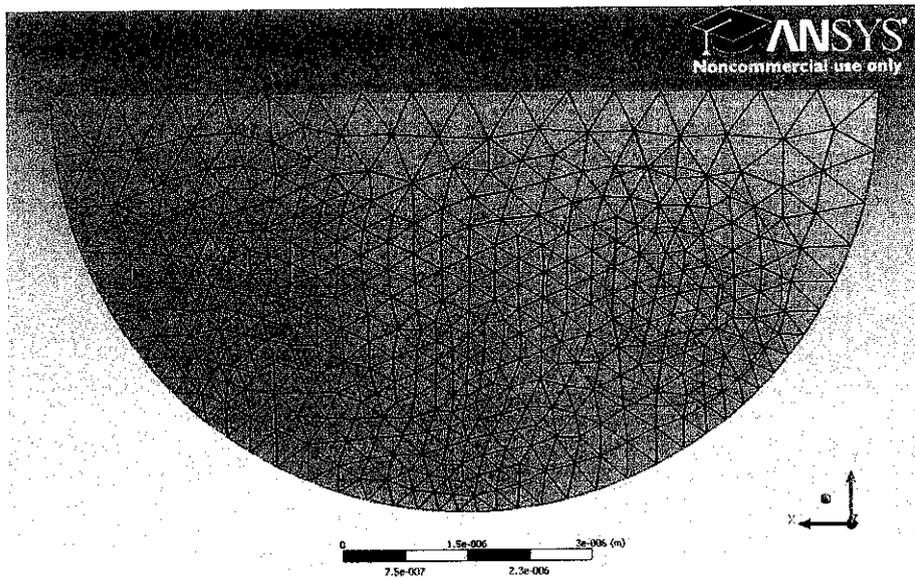
Outlet view

Isometric view

Figure 17 : Generated Mesh for Type A

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Type B generated mesh geometry for inlet, outlet and isometric view are showed in Figure 16.
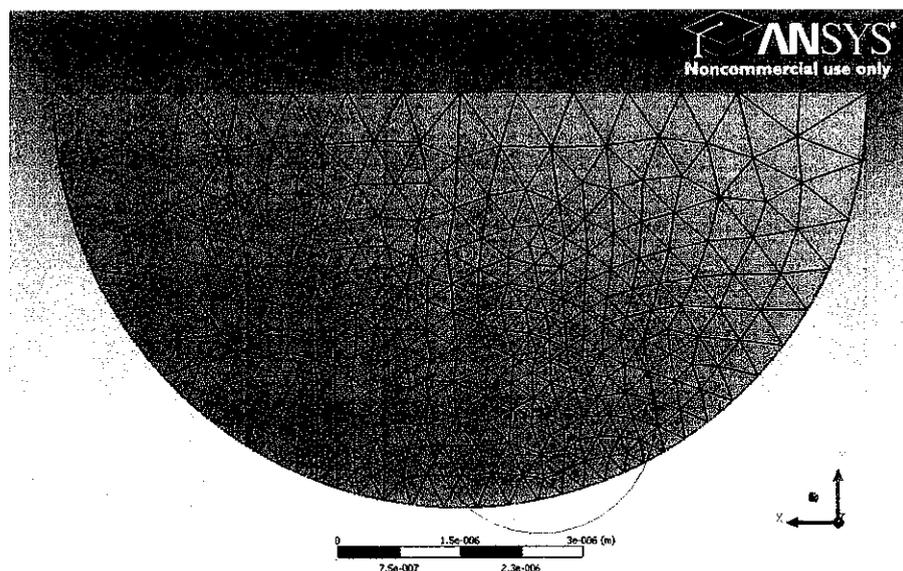
Inlet view

Outlet view
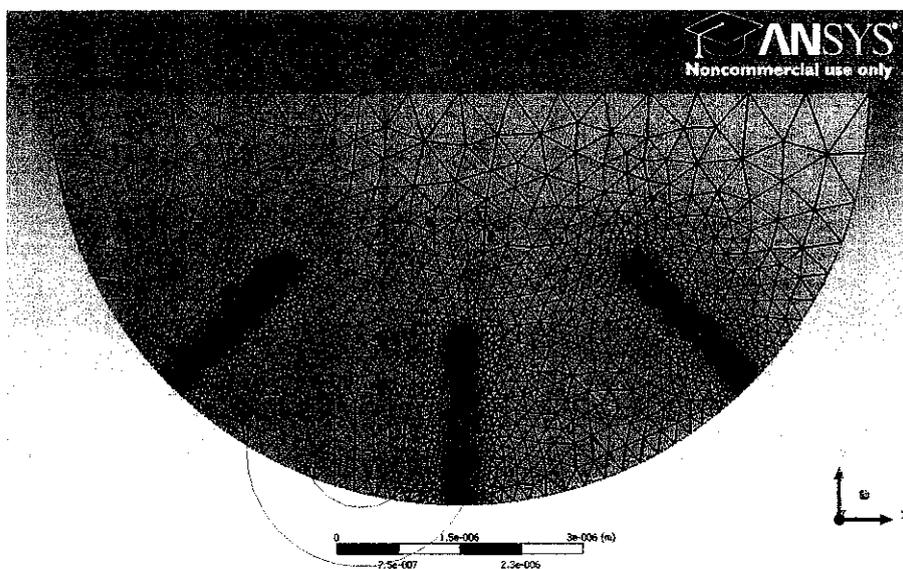
Isometric view
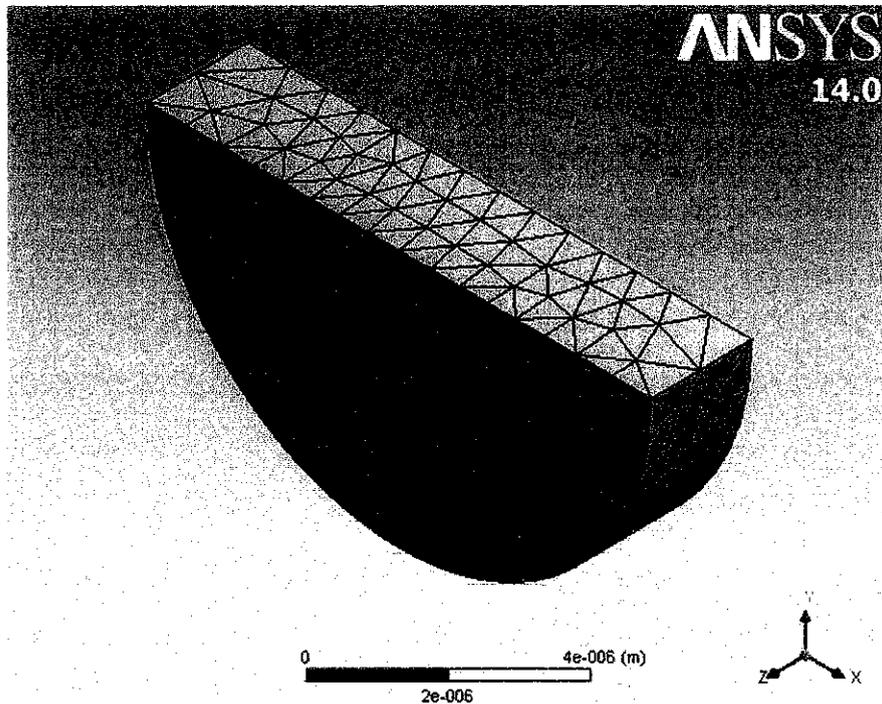
Figure 18 : Generated Mesh for Type B

3.1.3 SOLVER SETUP

In solver process, all the mathematical equations that need to be used along the simulation need to be determined. For example, in the case of study the flow along the pipeline, it is important to determine what are the type of the flow that will be used along the pipeline and the characteristics of the elements inside the pipeline such as density, molecular weight and diameter. By determine all the equations in the solver step, then all the calculation and results will be calculated in the post processing part.

3.1.3.1   Multiphase solver options

There are three types of models in the multiphase models which are mixture, volume of fluid and Euler. In this case, for Type A and Type B, both were using the Euler multiphase model approach. The interactions between the two phases in the

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system based on literature suggested that Eulerian model for multiphase flow would be an appropriate mathematical scheme to utilize for this type of two-phase flow. The volume fraction parameters in the Eulerian model options were specified to be explicit since the problem was said to be in a steady state.

Then, from the second parameters (velocity), laminar flow had been selected for the viscous option in solver setup. Laminar flow was specified for all models to be consistent with the 3 velocities given by OneBaja Project. Further calculation for the Reynolds number to determine the laminar or turbulence flow is showed. The calculation was made based on the highest velocity which is 706.65 m/s that was calculated from the volumetric flowrate given.

Reynolds number calculation:

$$Re = \frac{\rho V D}{\mu}$$

$$= \frac{609.945 \times 10^{-3} kg/m^3 \times 706.65 m/s \times 1 \times 10^{-6} m/s}{12.3705 \times 10^{-6}}$$

$$= 348 < 2300 \quad \therefore Laminar\ Flow$$

The materials were specified as nitrogen and hydrogen for every simulations performed. In phases, it was required to indicate the materials for the primary and secondary phase. The important to select the phase is because the secondary phase is the phase that will disperse throughout the continuous primary phase in the simulation. For this model, nitrogen was considered the primary phase while hydrogen was the secondary phase. The phase interactions were specified the Schiller-Naumann which is the default recommended in FLUENT for this multiphase model.

Next, boundary conditions for the zone were specified. All of the zones were remained as the default and only the inlet type was changed to the velocity-inlet. The velocity for each material (nitrogen and hydrogen) needs to be filled as the velocity that given by the OneBaja project but only in the secondary phase, the volume fraction was specified as 0.5. Figure 18 shows all the zones that available for the multiphase system.

```
zones,
    solid  (hydrogen)
    wall-solid  (hydrogen)
    interior-solid  (hydrogen)
    inlet  (hydrogen)
    outlet  (hydrogen)
    symmetry  (hydrogen)
    solid  (nitrogen)
    wall-solid  (nitrogen)
    interior-solid  (nitrogen)
    inlet  (nitrogen)
    outlet  (nitrogen)
    symmetry  (nitrogen)
    symmetry
    outlet
    inlet
    interior-solid
    wall-solid
    solid
Done.
```

Figure 19 : Available zones in multiphase system

In the solution method, Coupled scheme was selected for this multiphase problem. Coupled scheme is define as a form of two-way coupling for the primary and secondary phases where both of the phases will affect each other but the secondary phase particles did not interact. The solution initialization was changed to Hybrid Initialization in order to efficiently initialize the solution based on the simulation setup. The Courant number in the solution control was changed to 40 instead of 200 (default), and explicit relaxation factors were reduced to 0.4 for momentum and pressure to avoid divergence in the solution.

CHAPTER 4

RESULTS AND DISCUSSION

4.1 INTRODUCTION

The last step of CFD simulation is the post processing. This last step will be the most vital part where all the specific area of study will be combined with all the equations and come out with the results. The post processing is the step which the calculations will be made in order to solve all the equations and conditions that had been set in the pre processing and solver step. Then, from the results, the conclusion can be done.

Multiphase flow in a microchannel inside the microreactor was examined in order to determine program capabilities with multiple phases. As mention early, for this project only 2 μm of microchannel length and only symmetry (half of the cylinder) dimension was used to decrease the mesh size to meet the ANSYS limitation. Besides that, the decision was made because the amount of time spent on iterations since incorporating with two phases in the simulation was taxing on the program's computing capabilities.

The volumetric flowrate for nitrogen and hydrogen inside the microchannel was chose to be 3.33 ml/min which is equal to 706.65 m/s as default and it is stated to be a laminar flow. This velocity was chosen for consistency of the previous study so that no confusion about the velocity study will occur in the future. Nitrogen and hydrogen were also specified to have velocity of 2.22 ml/min which is equal to 471.10 m/s to see the effect of flow towards changes of velocity, besides to remain the flow in laminar flow regime. For some of the final simulations, nitrogen and hydrogen were specified to have velocity of 1.11 m/s which is equal to 235.50 m/s, in order to conclude the changes profile when different velocity parameters were provided.

The main objective of this project was to see the flow phenomena along the nanowires towards different configurations and velocities. The area that needs to be specified in this model was the nanowire that was located at the bottom of the outlet

face. Figure 19 shows the specified nanowire that need to be observed. Then, from the results, the volume fraction and the velocity profile along the nanowires inside the microchannel. In this multiphase problem, Eulerian models were tested in FLUENT with some results.
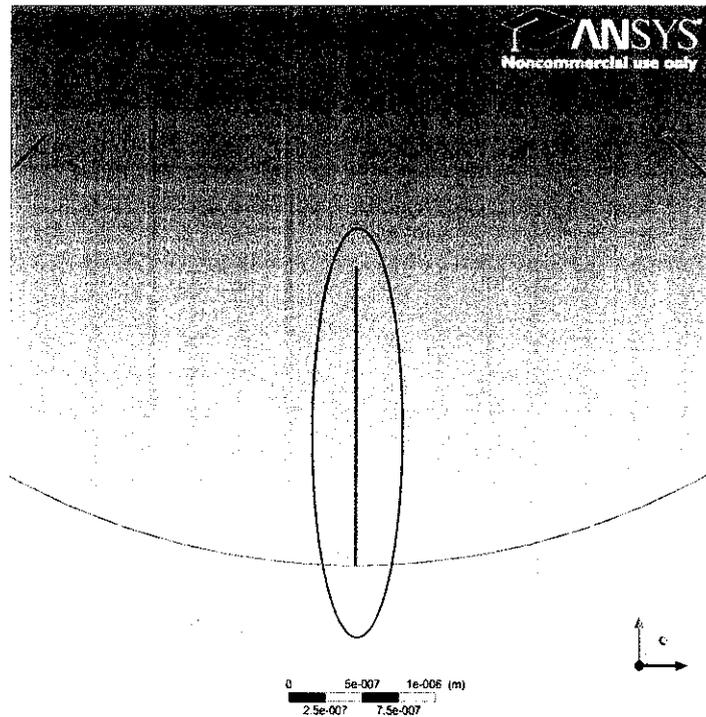
Figure 20 : Specified nanowire

4.2 CFD SIMULATION RESULTS

These tests were conducted to see the results for the volume fraction and the velocity profile for the specified nanowires. Details of each of these trials are shown and the final section will discussed the parameters effects that had been chose.

4.2.1 VOLUME FRACTION VS. VELOCITY

The significant study the volume fraction is to see at which point nitrogen and hydrogen have the highest number in terms of volume. From the results, it can be observed whether the configurations and dimensions for the nanowires along the microchannel is suitable with the velocity that had been introduced.

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4.2.1.1 Velocity of 706.65 m/s

Both of the configuration (Type A and Type B) were tested in FLUENT with some results. The simulated volume fraction results of nitrogen and hydrogen for Type A and Type B configuration can be seen in Figure 20 and Figure 21.
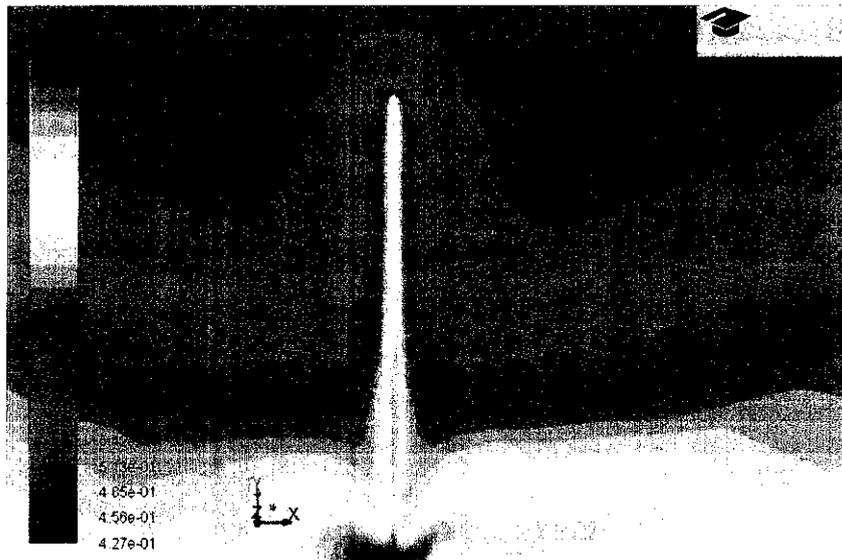
Contours of Volume fraction (nitrogen)   Aug 10, 2012
ANSYS FLUENT 14.0 (3d, dp, pbns, eulerian, lam)
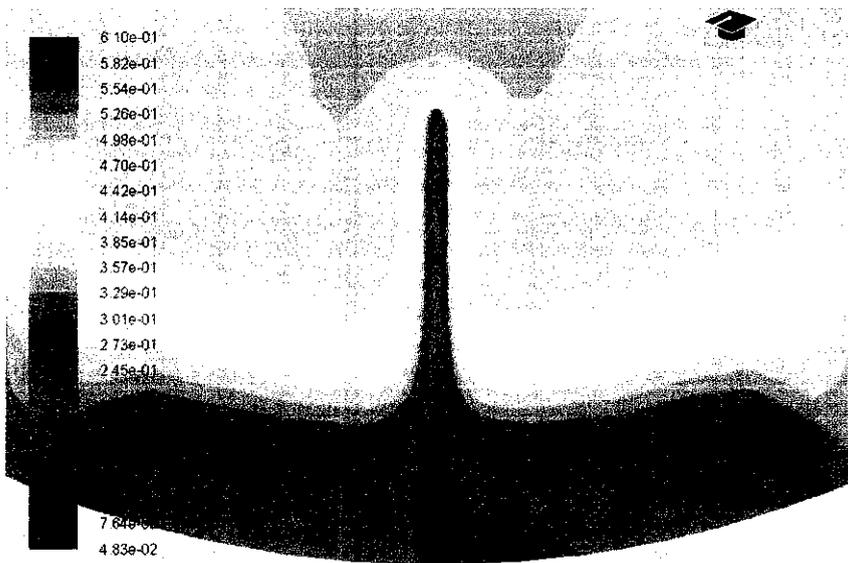
Contours of Volume fraction (hydrogen)   Aug 10, 2012
ANSYS FLUENT 14.0 (3d, dp, pbns, eulerian, lam)

Figure 21 : Volume fraction of nitrogen and hydrogen for Type A

Figure 22 : Volume fraction of nitrogen and hydrogen for Type B

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In the both Figure 20 and Figure 21, dark blue represents the lowest value of volume fraction and deep red represents the highest number of volume fraction. The phases can be observed to be separating at the specified nanowire whereby the nitrogen was tend have highest volume fraction along the nanowire while much of hydrogen was at the center of the microchannel. Based on the configuration, Type A was having small gradient of volume contour for both nitrogen and hydrogen whereby it indicates that there will be less of volume along the nanowires, compare to Type B. The scale from the contour shows the highest number of volume fraction for nitrogen and hydrogen is 0.998 and 0.573 for Type B, while 0.952 and 0.610 for Type A.

4.2.2   VELOCITY PROFILE VS. VOLUME FRACTION

Then, velocity contours was generated to see the velocity profile along the nanowire. The results shows that for both configurations, the velocity was becoming much slower at the nanowire compare to the center of the microchannel. Compare to Type B, Type A was plotted with lower value of velocity contour along the nanowire resulting the fluid tend to become slower when passing the Type A configuration. The highest velocity for nitrogen and hydrogen is 743 m/s and 1030 m/s for Type A, and 740 m/s and 1000 m/s for Type B.

Figure 22 and Figure 23 shows that the velocity profile results of nitrogen and hydrogen for Type A and Type B.
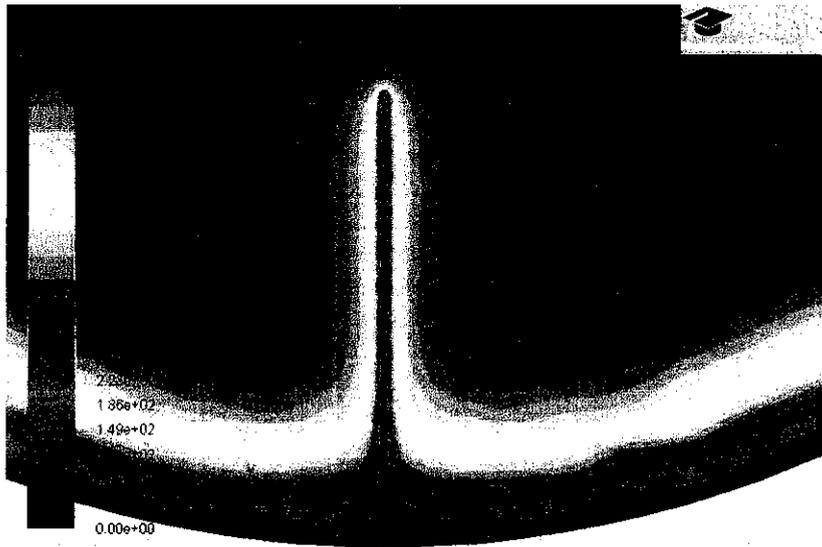
Contours of Velocity Magnitude (nitrogen) (m/s)

Aug 10, 2012
ANSYS FLUENT 14.0 (3d, dp, pbns, eulerian, lam)
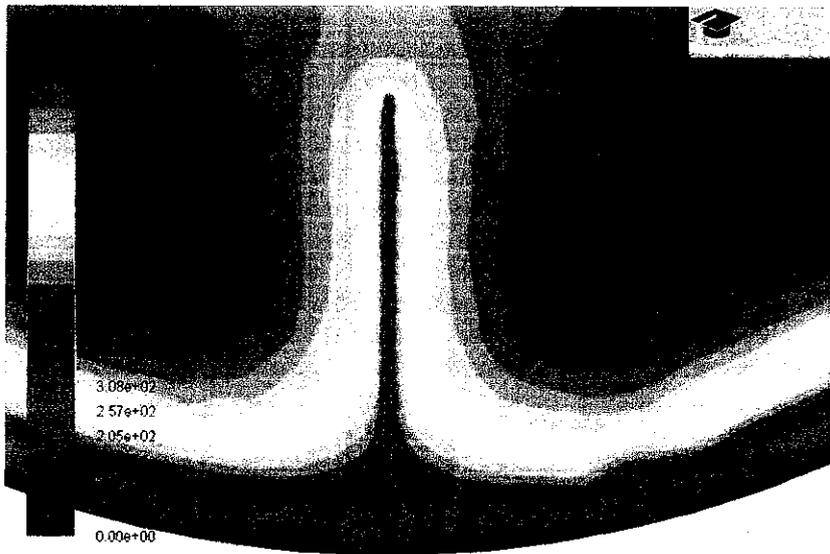
Contours of Velocity Magnitude (hydrogen) (m/s)

Aug 10, 2012
ANSYS FLUENT 14.0 (3d, dp, pbns, eulerian, lam)

Figure 23 : Velocity profile of nitrogen and hydrogen for Type A
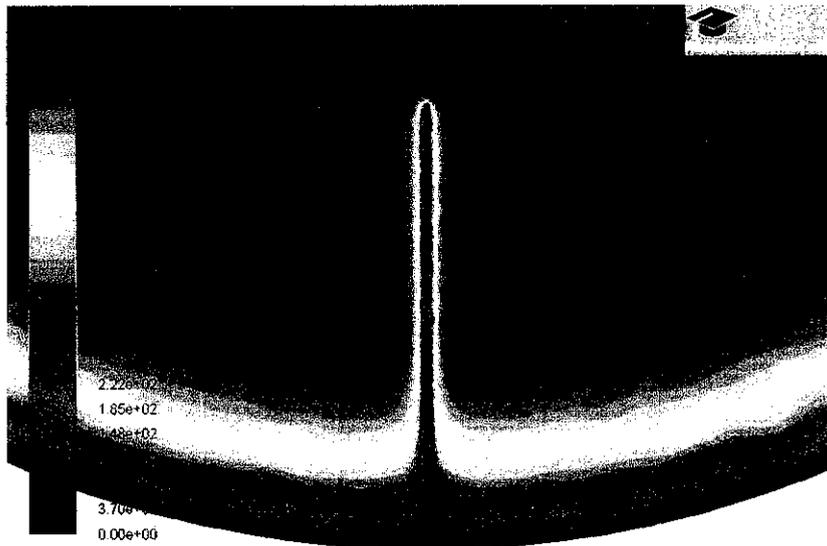
Contours of Velocity Magnitude (nitrogen) (m/s)
Aug 10, 2012
ANSYS FLUENT 14.0 (3d, dp, pbns, eulerian, lam)
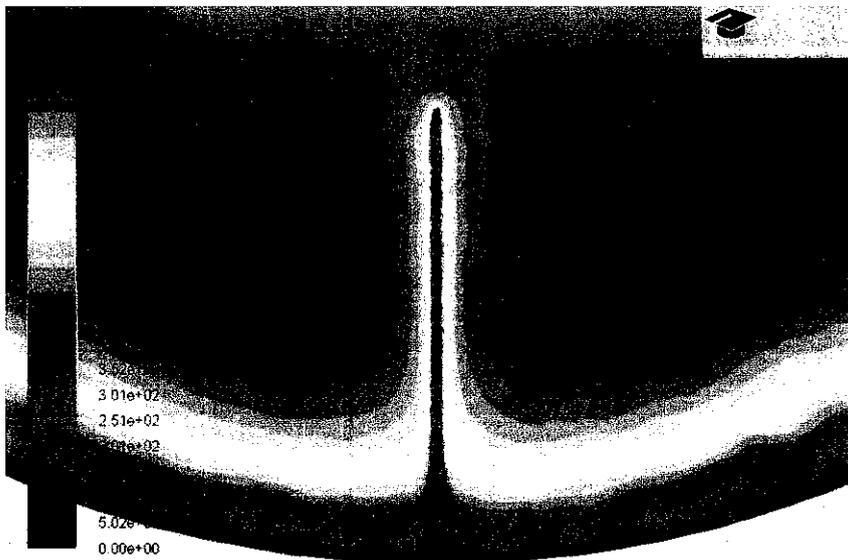
Contours of Velocity Magnitude (hydrogen) (m/s)
Aug 10, 2012
ANSYS FLUENT 14.0 (3d, dp, pbns, eulerian, lam)

Figure 24 : Velocity profile of nitrogen and hydrogen for Type B

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4.3   DISCUSSIONS

The contour study then repeated for the two velocities that had been introduced. The study was the same as the 706.65 m/s where the main objective to study the volume fraction and also the velocity profile. The setting for all setup in ANSYS 14.0 FLUENT was remained as the same. The changes had been made only at the inlet zone in boundary conditions. The inlet-velocity at the inlet zone for the nitrogen and hydrogen was changed to the next velocity, 471.10 m/s and then with 235.50 m/s for the third round. All of the 3 results had been compared to one of each others to see the volume fraction and velocity profile. The results for the volume fraction are shown in Table 2.

Type A

Velocity	Nitrogen		Hydrogen	
	Max	Min	Max	Min
706.65 m/s	0.99800	0.42700	0.57300	0.00212
471.10 m/s	0.99800	0.33300	0.66700	0.00241
235.50 m/s	0.99100	0.21500	0.78500	0.00896

Type B

Velocity	Nitrogen		Hydrogen	
	Max	Min	Max	Min
706.65 m/s	0.95200	0.39000	0.61000	0.04830
471.10 m/s	0.93400	0.38600	0.61400	0.06590
235.50 m/s	0.93000	0.37900	0.62100	0.06970

Table 2 : Minimum and maximum volume fraction

Next, for the velocity profile inside the microchannel, the results for the minimum and maximum value of velocity flowing through the nanowires can be shown in Table 3.

Type A

Velocity	Nitrogen (m/s)		Hydrogen (m/s)	
	Max	Min	Max	Min
706.65 m/s	743.00	0	1030.00	0
471.10 m/s	500.00	0	712.00	0
235.50 m/s	256.00	0	376.00	0

Type B

Velocity	Nitrogen		Hydrogen	
	Max	Min	Max	Min
706.65 m/s	740.00	0	1000.00	0
471.10 m/s	498.00	0	690.00	0
235.50 m/s	254.00	0	359.00	0

Table 3 : Maximum and minimum velocity profile

Based on the results obtained from the ANSYS, there are a few discussions that can be stated here. First, the characteristic of nitrogen and hydrogen towards the configurations and velocity can be analyzed. Then, the study on how the type of flow inside the microchannel will affect the movement of nitrogen and hydrogen. The results showed that the velocity becoming much slower along the nanowire in any configurations. The decision to study the nanowires inside the microchannel is acceptable since to build the microreactor will consume a lot of time and money. Simulate the flow characteristic using any simulation programs seems to be right compare jump into the experiment from the start. The results then can be used to build the microreactor according to the suggestions from the simulation.

Then, for the volume fraction along the nanowires, the result obtained was not very unsatisfied since there are few modifications that need to be done in the future. The results shows that the nitrogen was having much volume along the nanowire compare to hydrogen whereby, most of nitrogen was flowing through the center of the

microchannel. In conclusion, the nitrogen will react with only a few of hydrogen along the nanowires where the catalyst is located. This will not contribute to the better production of ammonia but the production will becoming lower than the convention method.

The results can be illustrated more from the point surface that had been specified as Figure 25. The details of location for the surface point had been showed in the next figure, Figure 26. The value of volume fraction and velocity at the specified point then will be analyzed.
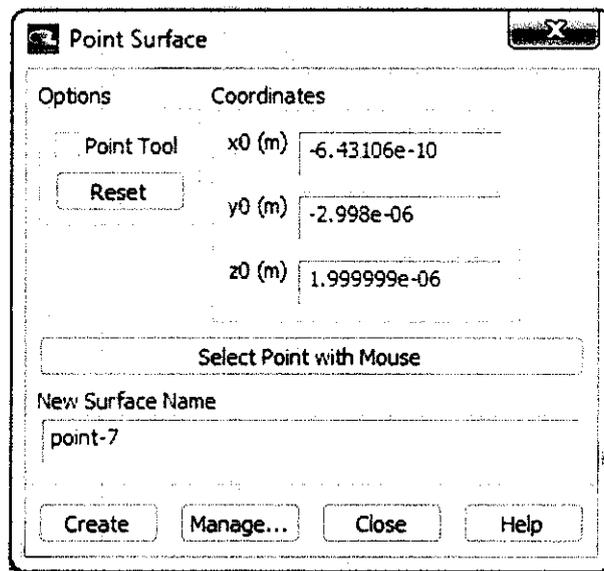
Figure 25 : Point surface
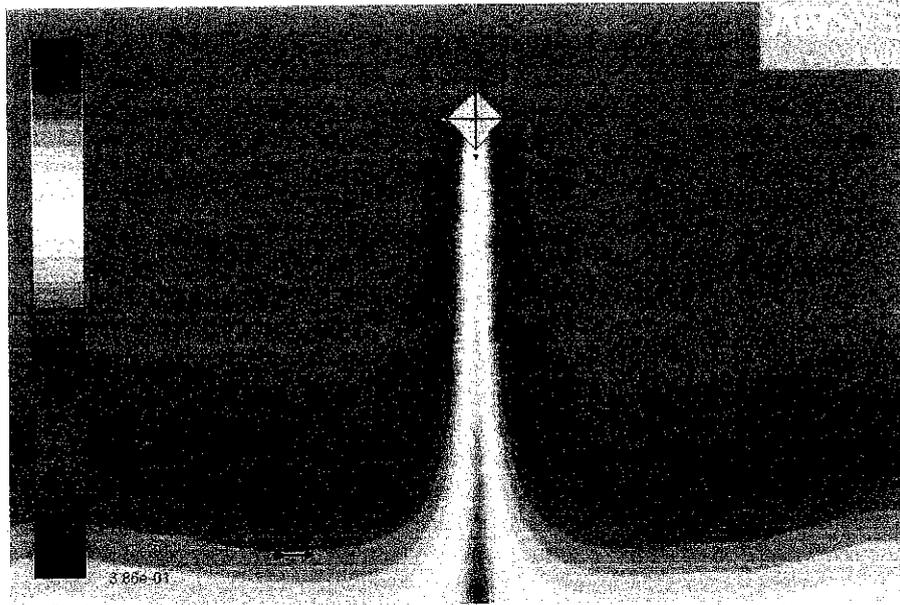
Contours of Volume fraction (nitrogen)

Aug 12, 2012
ANSYS FLUENT 14.0 (3d, dp, pbns, eulerian, lam)
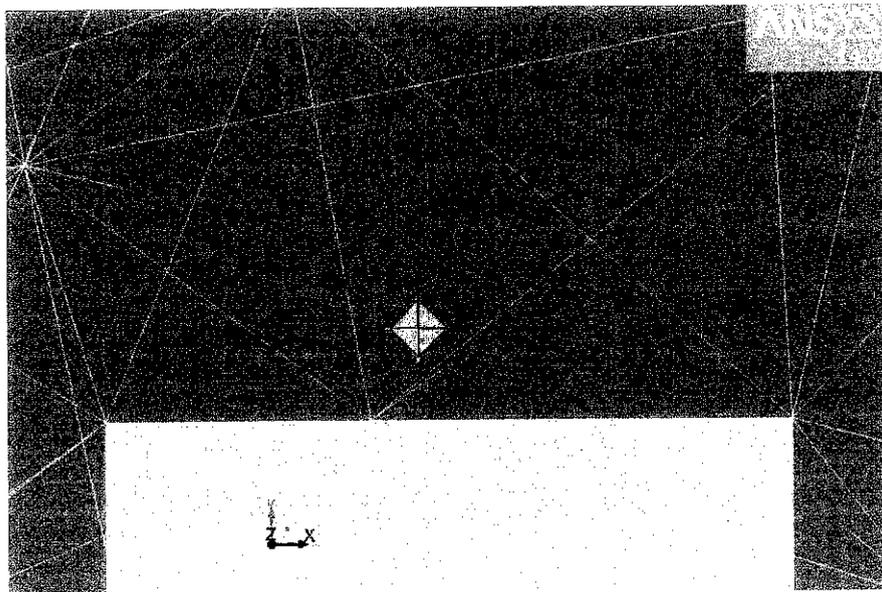
Highlight: Surfaces

Aug 12, 2012
ANSYS FLUENT 14.0 (3d, dp, pbns, eulerian, lam)

Figure 26 : Details of the location for point surface

From the surface point that had been specified earlier, the results for the volume fraction and velocity profile are shown in the Table 4. Then, from the results, three graphs of the velocity profile (nitrogen) vs. velocity profile (hydrogen), volume fraction vs. velocity, and velocity profile vs. velocity, at the point surface, are shown in the Figure 27, Figure 28 and Figure 29.

Velocity	Type	Volume Fraction		Velocity Profile	
		Nitrogen	Hydrogen	Nitrogen	Hydrogen
235.5	Type A	0.723	0.277	20.6	33.7
	Type B	0.703	0.297	23.8	40.2
471.1	Type A	0.754	0.246	43.3	68.2
	Type B	0.718	0.282	50.1	81.3
706.65	Type A	0.771	0.229	68.9	102
	Type B	0.730	0.270	78.2	122

Table 4 : Specified surface point results for volume fraction and velocity profile

Figure 27 and Figure 28 shows the velocity profile at the point surface for nitrogen and hydrogen. From the both graphs, it can be conclude that nitrogen had high velocity compare to hydrogen for both configurations. This is because of the characteristics of nitrogen that have high molecular weight of 28 u compare to hydrogen, 6 u (from the equation). The results also shows that nitrogen was tend to fill the bottom part while hydrogen was at the center of the microchannel.

Then, from the Figure 29 and Figure 30, the graphs shows the results of volume fraction and the velocity profile for nitrogen since the velocity profile for nitrogen vs. hydrogen had been approved in Figure 27 and Figure 28. Secondly, the reason why the graphs only show the nitrogen only is because, for the volume fraction, the total of volume fraction must be equal to 1. So, it is enough to study only one element to save time. From the graph, it can be said that Type A shows that the volume fraction is high while the velocity is low compare to Type B near the nanowire.
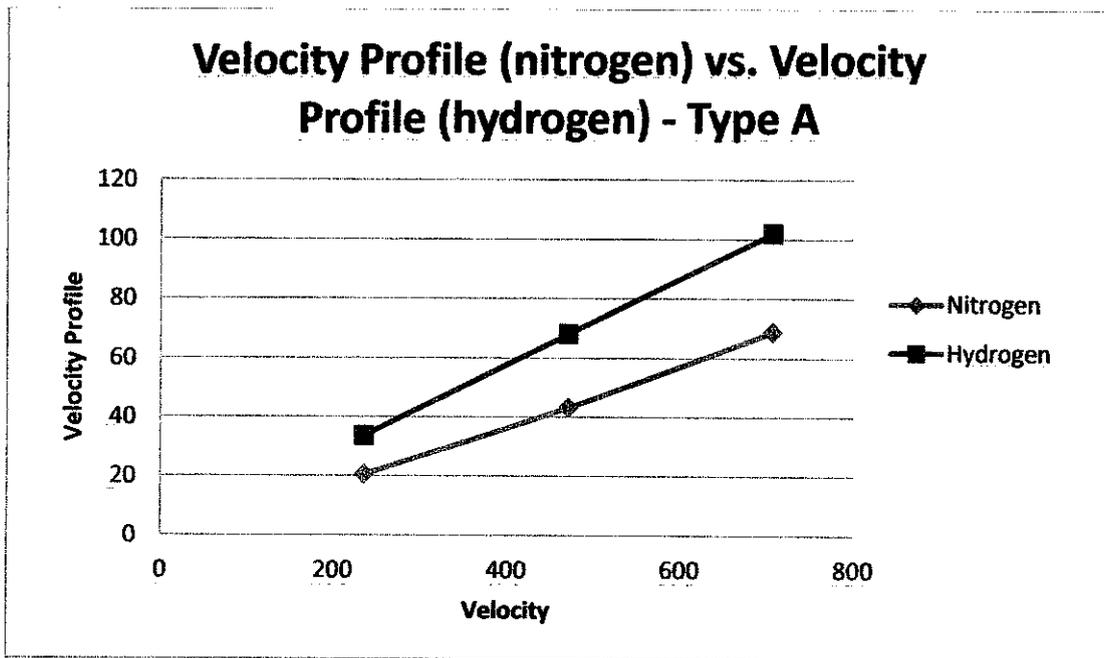
Figure 27 : Nitrogen vs. Hydrogen for Type A configuration
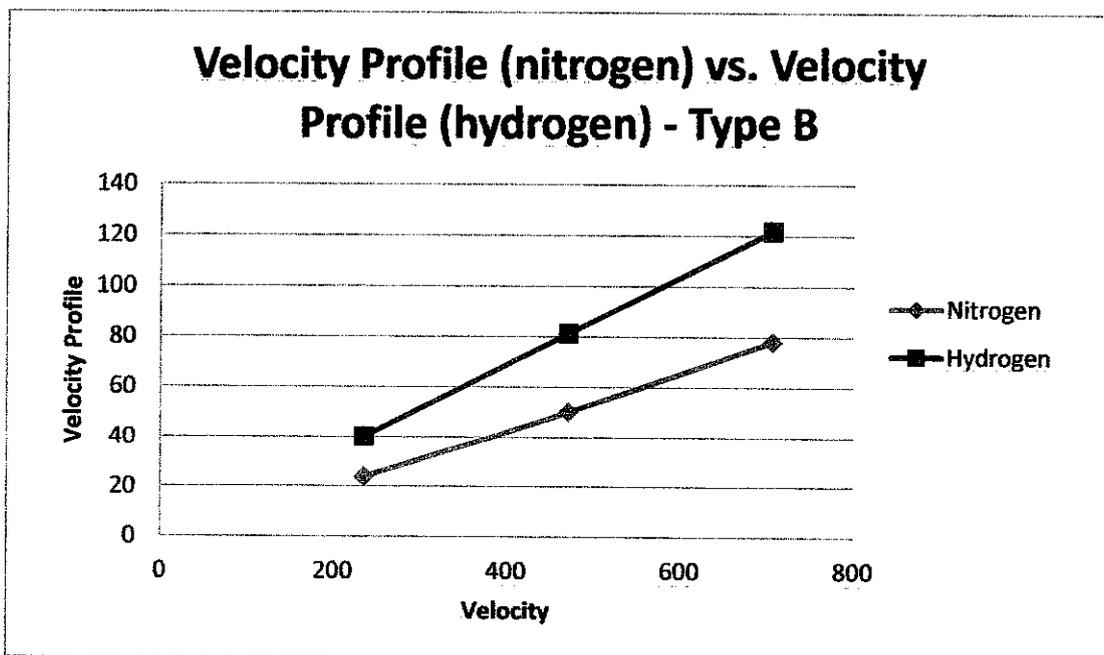
Figure 28 : Nitrogen vs. Hydrogen for Type B configuration
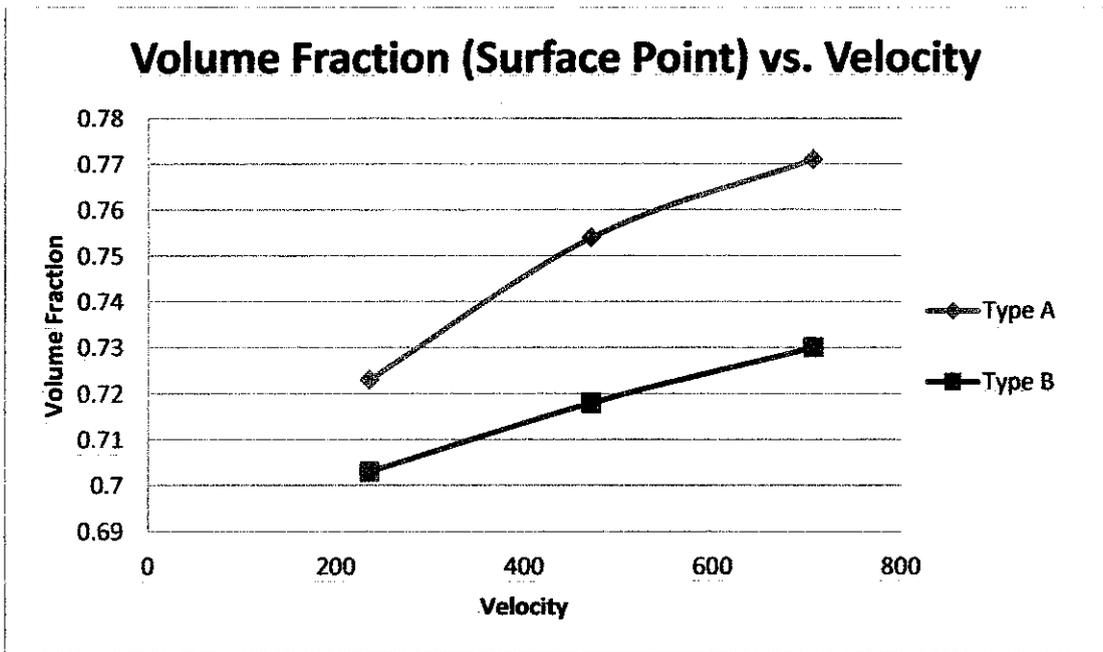
Figure 29 : Volume fraction vs. velocity at the surface point for nitrogen
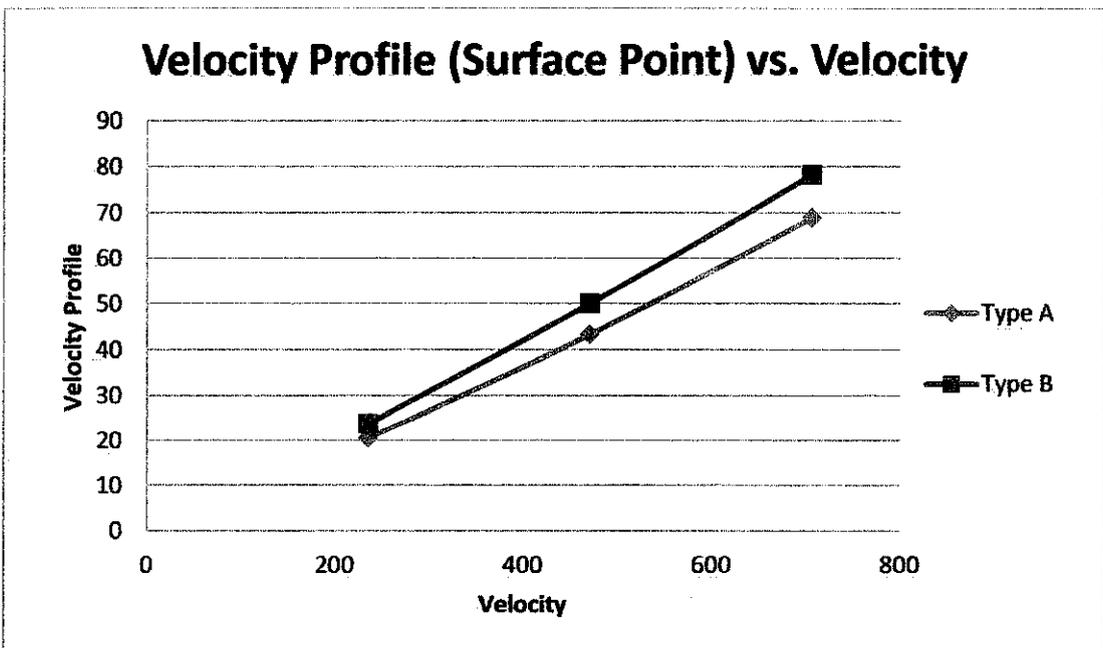
Figure 30 : Velocity profile vs. velocity at the surface point for nitrogen

CHAPTER 5

CONCLUSIONS & RECOMMODATIONS

5.1   CONCLUSIONS

As the conclusion, the main objective for the project to computationally design the microreactor had been archived. Although the microreactor only can be done for 2 μm of length due to the software limitation, further improvement on the design might be needed before build the microreactor. By doing the simulation, most of the specification for the microreator can be done and it will save lots of money.

The effect of nanowires configuration also had been archived with the specification that had been introduced by OneBaja project. Further improvement in ANSYS FLUENT setup is needed in order to see the most effective reaction site along the nanowires. The results show that nitrogen and hydrogen was not gave the right volume fraction of 0.25 of nitrogen and 0.75 of hydrogen along the nanowire in order to have the optimum reaction to produce ammonia (from the equation). Although the result shows that, Type A might be preferable compare to Type B because it has slower velocity, but the volume fraction for the nitrogen and hydrogen near the nanowire is not right based on the stoichiometry number.

There are a few reasons that lead to this phenomenon. One of them is molecular weight of the molecule. Nitrogen (28 u) is heavier than hydrogen (6 u) which resulting nitrogen to favor to go to the bottom side of microchannel. Secondly, the main reason for this phenomenon is because the type of flow inside the microchannel. The flow is laminar instead of turbulence which is a steady flow that will affect the distribution of reactant gas flow inside the microchannel.

5.2 FUTURE WORKS

Lastly, there are things need to be improved for further study in order to have a better production of ammonia:

1. To study the effect of turbulence flow inside the microchannel

2. To have more nanowires configurations in order to have better volume fraction and velocity profile.

3. To change the length of nanowires (within 5 μm) in order to see the effect of turbulence.

4. To change the width of microchannel from 2 μm to 10 μm to see the effects of flow on the nanowires more clearly.

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APPENDICES

APPENDIX 1-1: "FINE" MESHING

Although there are few limitation using the ANSYS with teaching license in FLUENT, the mesh geometry still can be generated and further improvement might be come out with better solutions. Type A generated mesh geometry with "fine" relevance center setup for inlet, outlet and isometric view are showed in Figure 14.

Inlet view
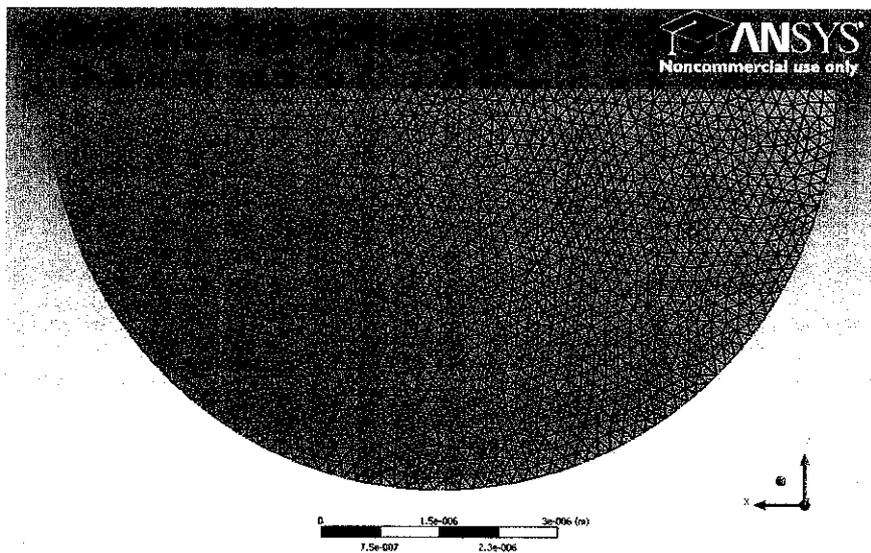
Outlet view
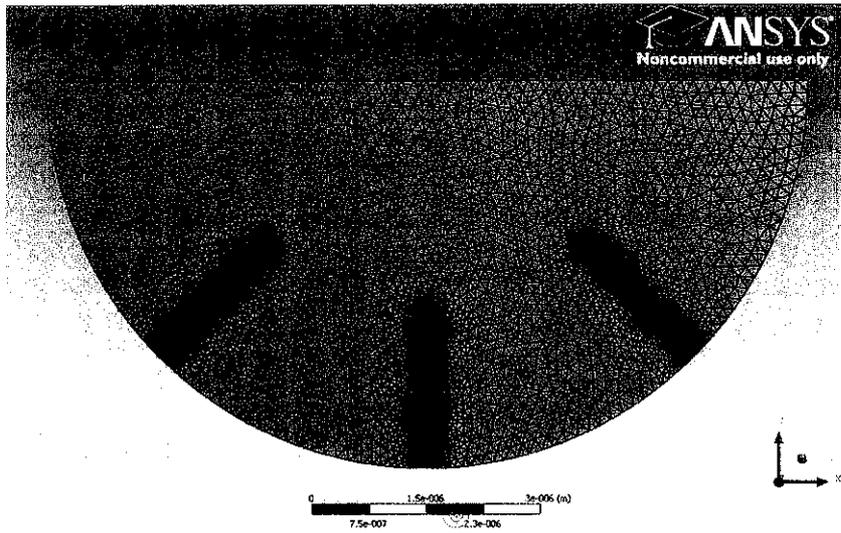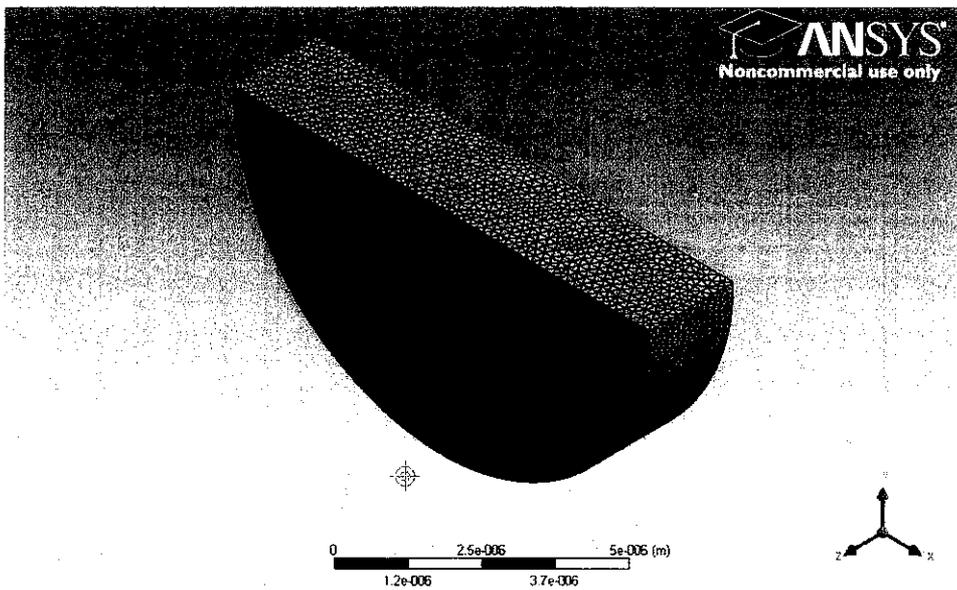
Isometric view

Figure 31 : Generated Mesh for Type B

Type B generated mesh geometry with "fine" relevance center setup for inlet, outlet and isometric view are showed in Figure 15.

Inlet view
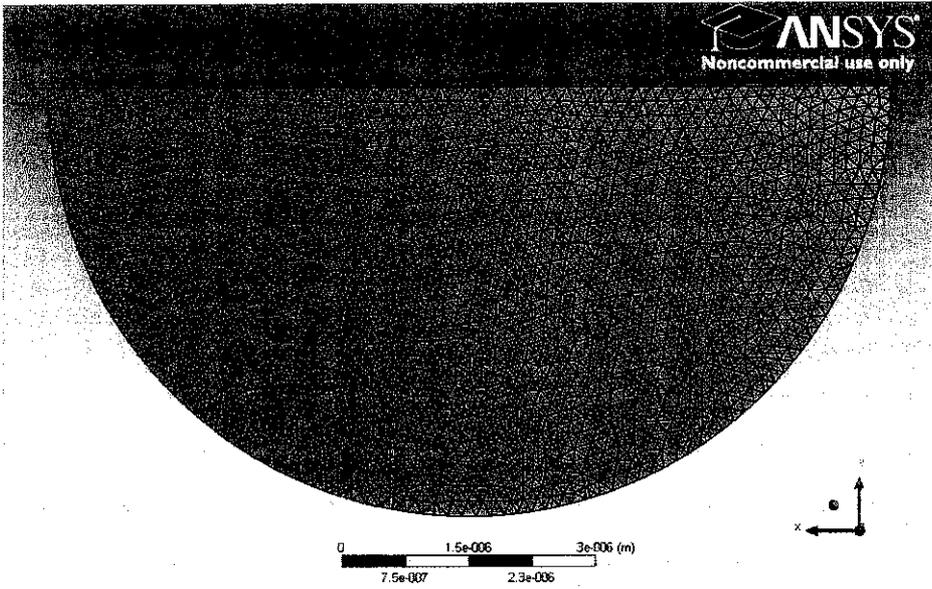
Outlet view
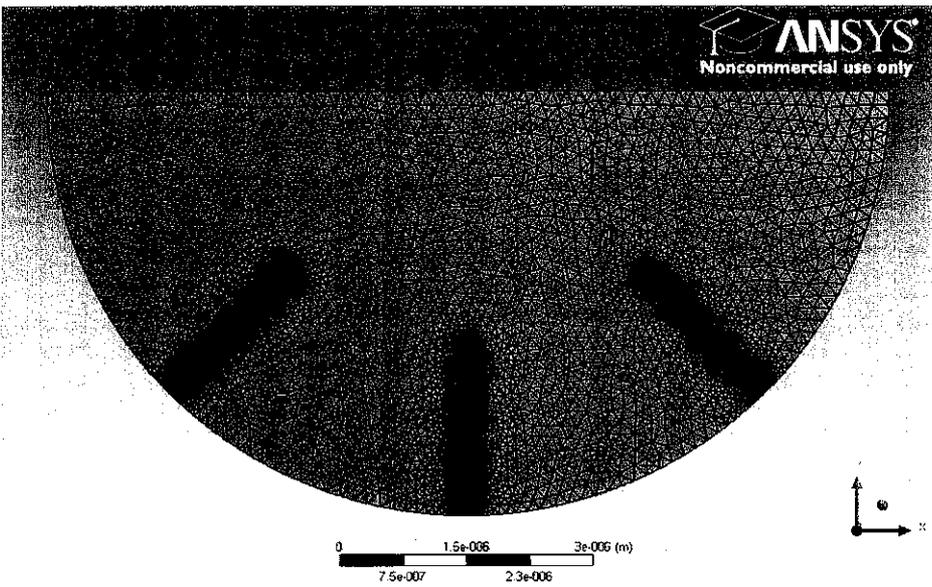
Isometric view
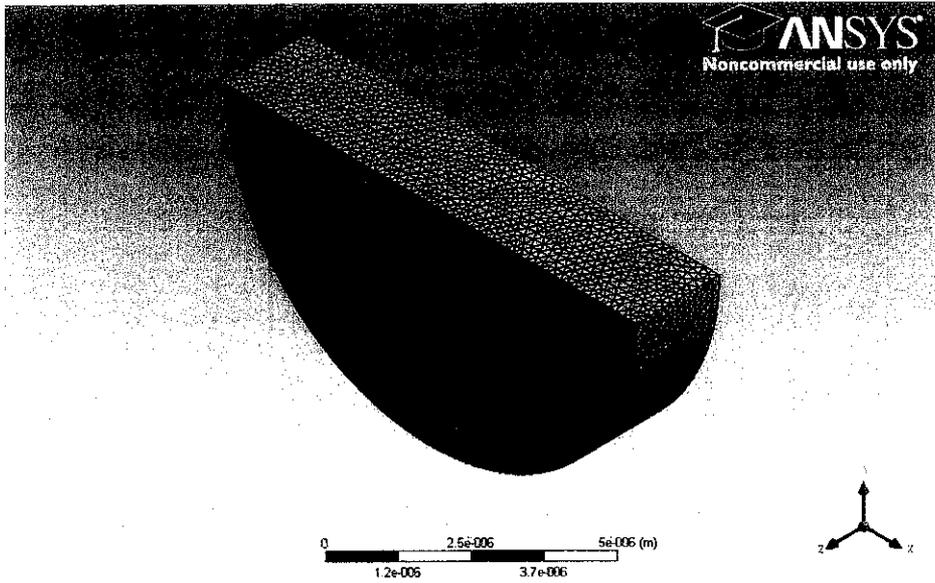
Figure 32 : Generated Mesh for Type B