

# 06-663 S20 Project Report: Modeling of a Tubular Reactor with a Non-Isothermal Cooling Jacket

Team 9

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## Abstract

In the modern chemical industry, some exothermic reactions take place in tubular reactors with non-isothermal cooling jackets, which involve a complex reactor design. The design considers various elements including tube sizing, coolant temperature and higher order reactions. This report considers the impact of these parameters on the design of a tubular reactor with a non-isothermal cooling jacket. The reaction studied is an exothermic and irreversible reaction in a tubular reactor; the reaction is assumed to be in liquid phase and the laminar flow regime. It is also assumed that no back mixing occurs in the reactor system. The reactor temperature is kept cool via a cooling jacket. Both elementary and second order reactions are studied. The reaction  $A + B \rightarrow C$  occurs inside the tube of a plug-flow tubular reactor. The Specific reaction studied is the propylene glycol production:

*Propylene oxide (A) + Water (B)  $\rightarrow$  Propylene Glycol (C)*. The simulation of the tubular reactor is performed in COMSOL<sup>[1]</sup>. The model setup is obtained from the COMSOL Application Gallery to ensure the reliability of the model setup. The Geometry used for the model is a 2D Axisymmetric Space Dimension. Physics considered are Transport of Diluted Species, Reaction Engineering, Heat Transfer in Fluids and Coefficient Form Boundary PDF.

This report presents a discussion of an optimal steady-state tubular reactor design, considering the impact of parameters including coolant conditions, inlet stream's flow rate, temperature and concentration, and reactor sizing. First, the introduction presents a brief description on the use of tubular reactors in both industries and research studies. Then, the geometry setup, mathematical modeling, and Numerical Methodology are presented accordingly. Next, the results along both axial and radial distance of the tubular reactor are presented and analyzed. We find that a longer reactor will result in higher conversion at the outlet. Decreasing the flow rate, concentration, and ratio of propylene oxide to water can also help increase the conversion at the outlet of the reactor. Finally, comprehensive suggestions on optimal design on the tubular reactor with a non-isothermal cooling jacket will be given.

## Introduction

Tubular reactors are used extensively in the chemical and biotechnology industry; they have also been widely studied by researchers in the field of chemical engineering. Tubular reactors usually consist of a cylindrical pipe with openings on each end for reactants and products to flow through. In industry, tubular reactors are usually operated at steady-state; and reactants are continually consumed as they flow down the length of the reactor. As the reactants flow down the reactor and the reaction takes place, an axial concentration gradient is formed, with products and unreacted reactants flow out of the reactor continuously.

Tubular reactors are a popular choice for the chemical production industry, especially in gasoline production, oil cracking, synthesis of ammonia from its elements, and the oxidation of sulfur dioxide to sulfur trioxide<sup>[2]</sup>. It is preferable to use tubular reactors in industrial applications due to several advantages. Most importantly, tubular reactors require easy maintenance because they are mechanically simple and easy to clean. There are no moving parts in a tubular reactor. Additionally, tubular reactors are good for large capacity processes and with low pressure drops, reducing the risks for runaways to a large extent. Tubular reactors can also be used bio-product manufacturing for small scale production, such as the production of algae and colloidal lignin. In the study of colloidal lignin<sup>[3]</sup>, the use of tubular reactors in biotechnology is gaining attention since it ensures better colloidal dispersions within the reactor compared to a stirred mixer reactor. The tubular reactor also enables researchers to scale up the reaction to production plant applications.

Since tubular reactors are used in various different applications, researchers are interested in phenomena in a tubular reactor under different reaction conditions. Luyben<sup>[4]</sup> studied the optimal steady-state economic design of cooled tubular reactor systems, considering an exothermic, irreversible, gas-phase reaction  $A + B \rightarrow C$  occurring. The study shows that for systems with small specific reaction rates the optimum design requires large reactors and large recycle flow rates and therefore small pressure drops. As the specific reaction rate increases, reactor size of

the recycle flow decreases; the optimum design then requires smaller tube diameters and larger pressure drops.

Similar to Luyben's study, in this project a tubular reactor with internal cooling is studied. However, the specific reaction considered is an exothermic, irreversible, liquid-phase reaction. Particularly, we are interested in the temperature and conversion profiles in a tubular reactor under elementary and higher order reactions, varying inlet flow and coolant conditions, and different reactor sizing.

### Geometry Setup

COMSOL Multiphysics is used to model the tubular reactor with a cooling jacket. A 2D Axisymmetric Space Dimension is used to study the behavior of this reactor along the axial and radial direction.

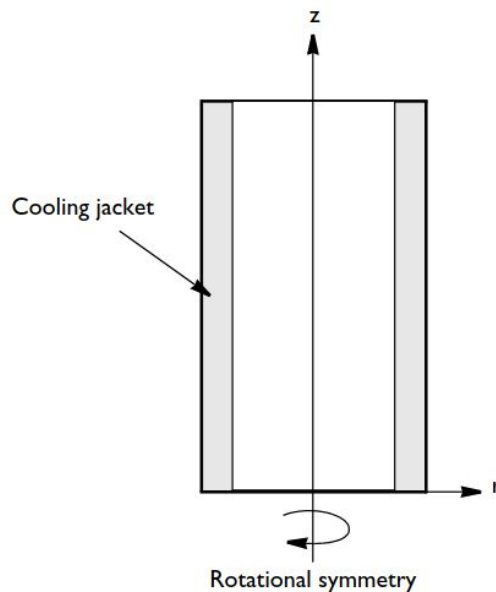


Figure 1: Schematic of the Tubular Reactor Geometry to be Modeled

Four major Physics are used in the study of the behavior of the reactor, subject to certain conditions. These are:

1. Transport of Diluted Species (tds)

As the reaction studied is a pseudo-first order reaction with respect to propylene oxide, the concentration of water is assumed to be constant. The rate law is thus:  $r_a = -k * C_a$ , where  $C_a$  is the concentration of propylene oxide. The tds module is thus used in case of the pseudo-first order reaction.

## 2. Reaction Engineering

The temperature and concentration profiles for the second order reaction are also studied, wherein the rate law considered for the reaction  $A + B \rightarrow C$  is:

$r_a = -k * C_a C_b$ , where a and b are propylene oxide and water. The reaction engineering is directly incorporated into COMSOL as a replacement of the tds module.

## 3. Heat Transfer in Fluids

The heat transfer in fluids module is used to simulate temperature profiles along the radius and length of the reactor, since it is operated under non-isothermal conditions.

## 4. Coefficient Form Boundary PDF

The boundary condition is used to model the temperature profile of the cooling jacket.

The dependant variable is set as  $T_j$ , with the dependent variable quantity unit set to K and the source term quantity unit to W/m, to be consistent with the constant term units in the differential equation.

The model is then run at steady state with the Physics incorporated as per the type of reaction. A rectangular geometry is used with a dimension of width at 0.1m and length at 1m. Next, parametric sweeps are done along the geometry. A standard mesh size of mapped distribution is used with the following attributes:

1. Across the inlet and outlet, the number of elements is set to 50, with an element ratio of 0.01, and a Geometric sequence growth and a flow in reverse direction.
2. Across the radial direction, the number of elements is set to 200, with an element ratio of 0.01, and a Geometric sequence growth and a flow in reverse direction.

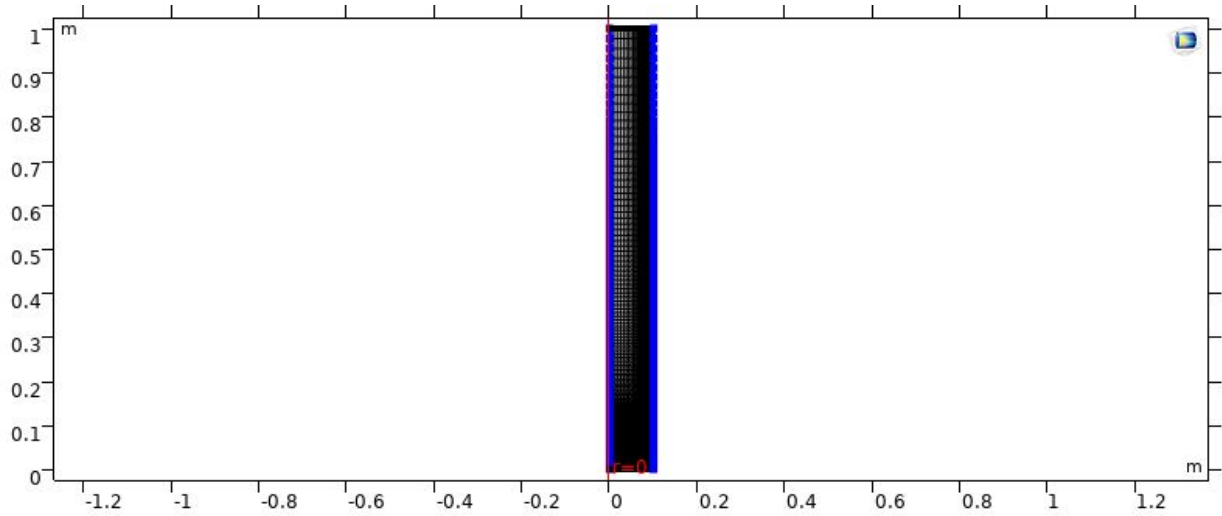


Figure 2: A general mesh representation for width of 0.1m and length of 1m.

## Mathematical Modeling

### i) Assumption

1. Steady State
2. Newtonian Fluid: incompressible, with physical properties of constant density  $\rho$ , viscosity  $\mu$ , thermal conductivity  $k$ , and diffusion coefficient  $D_A$ .
3. All species with same diffusivity
4. Homogeneous, irreversible reaction
5. Laminar flow in axial direction, no rotation and radial velocity
6. Diluted species, no back mixing

### ii) Governing Equations

- Equation of continuity:

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

- Momentum balance:

$$\rho \frac{D\mathbf{v}}{Dt} = -\nabla p - \nabla \cdot \boldsymbol{\tau} + \rho \mathbf{g}$$

- Energy balance:

$$\frac{\partial}{\partial t} \rho \left( \hat{U} + \frac{1}{2} \mathbf{v}^2 \right) = -\nabla \cdot \mathbf{e} + \rho (\mathbf{v} \cdot \mathbf{g})$$

- Mass balance:

$$\frac{\partial}{\partial t} \rho_A = -\nabla \cdot n_A + r_A$$

### iii) Constitution Laws

Under our assumption and combining the momentum balance and the equation of continuity, the fluid velocity in the tubular reactor can be expressed as the Poiseuille flow. The flow is only in  $z$  direction and depends on radius  $r$ , which gives us momentum transport properties. For heat transfer, since constant density is assumed throughout the reactor, the energy balance can be simplified by the Fourier's law of heat conduction. Finally, for mass transport, we model diffusion of each component by the Fick's law. The equations of the constitutive laws are given below:

- Poiseuille flow:

$$U(r) = U_0 \left(1 - \left(\frac{r}{R}\right)^2\right)$$

Where  $U(r)$  is the velocity in  $z$  direction,  $U_0$  is inlet velocity at the center of the reactor and  $R$  is the radius of the reactor.

- Fourier's Law of Heat Conduction:

$$q = -k \nabla T$$

Where  $q$  is the heat flux,  $k$  is the thermal conductivity, and  $\nabla T$  is the temperature gradient.

- Fick's Law:

$$j_A = -\rho D_A \nabla \omega_A$$

Where  $j_A$  is the diffusion flux,  $D_A$  is the diffusion coefficient and  $\nabla \omega_A$  the mass fraction for component A.

#### iv) Model Formulation

From the previous section, we can formulate our model by three balanced systems: mass balance of chemical reactions, energy balance in the reactor, and energy balance in the cooling jacket.

- Mass balance (equation of continuity for species A) in cylindrical coordinates:

$$D_A \left( \frac{1}{r} \frac{\partial C_A}{\partial r} + \frac{\partial^2 C_A}{\partial r^2} + \frac{\partial^2 C_A}{\partial z^2} \right) - 2U_0 \left( 1 - \left( \frac{r}{R} \right)^2 \right) \frac{\partial C_A}{\partial z} + r_A = 0$$

Where  $C_A$  denotes the concentration of species A and  $r_A$  is the reaction rate.

##### Boundary Conditions:

- Inlet ( $z = 0$ )

$$C_A(r, 0) = C_{A0}$$

- At wall ( $r = R$ )

$$\frac{\partial C_A}{\partial r}(R, z) = 0$$

- Outlet ( $z = L$ )

$$\frac{\partial C_A}{\partial z}(r, L) = 0$$

Where  $L$  is the length of the reactor.

The first BC gives inlet concentration  $C_{A0}$ . The second BC states that there is no diffusion flux at the wall of the reactor. The third BC states that transport at the outlet is dominated by convection over diffusion.

- Energy balance for Newtonian fluids in cylindrical coordinates:

$$k \left( \frac{1}{r} \frac{\partial T}{\partial r} + \frac{\partial^2 T}{\partial r^2} + \frac{\partial^2 T}{\partial z^2} \right) - 2U_0 \left( 1 - \left( \frac{r}{R} \right)^2 \right) \rho C_P \frac{\partial T}{\partial z} - r_A (-\Delta H_{RX}) = 0$$

Where  $T$  is temperature,  $C_P$  is heat capacity  $\Delta H_{RX}$  is reaction enthalpy

##### Boundary Conditions:

- Inlet ( $z = 0$ )



$$T(r, 0) = T_0$$

- Wall (  $r = R$  )

$$-\frac{\partial T}{\partial r}(R, z) = \frac{U_k}{k}(T - T_a)$$

- Outlet (  $z = L$  )

$$-\frac{\partial T}{\partial z}(r, L) = 0$$

Where  $T_0$  denotes inlet temperature,  $U_k$  is the heat transfer coefficient between the reactor and cooling jacket.

Similar to mass balance, there are three boundary conditions at inlet, wall, and outlet. The first BC is the temperature of feed at inlet, the second BC is heat exchange between reactor and cooling jack, and the third BC states that outflow heat is purely convective.

- Energy balance in cooling jacket

$$\frac{\partial T_j}{\partial z} = \frac{2\pi R U_k (T - T_j)}{m_c C_{Pc}}$$

Where  $T_j$  is the coolant temperature,  $m_c$  is the mass flow rate of coolant,  $C_{Pc}$  is the heat capacity of coolant. We assume heat exchange between the jacket and reactor only through convection, so conduction and radiation effects can be neglected.

Boundary Condition:

- Inlet (  $z = 0$  )

$$T_j(0) = T_{a0}$$

Where  $T_{a0}$  is the coolant temperature at jacket inlet.

## Numerical Methodology

Based on the model of propylene glycol production in the tubular reactor, we want to maximize the conversion of both the first order and second order reaction at the outlet. Different parameters are varied respectively to find the optimal result. The size of the reactor, coolant fluid

temperature, coolant fluid direction, flow rate of reactant and inlet concentration of the reactant for both the first order and second order reaction considered in this project. Also, the temperature profiles inside the reactor are within the scope of this project to guarantee the safety of production.

Our model is a classical non-isothermal plug flow reactor (PFR) and we are familiar with characteristics of this reactor from semester's courses. In our expectations, the rate of reaction of the second order reaction will be much faster than that of the first order reaction; and we need to ensure our heat exchange device does not allow the temperature inside the reactor to increase too much. Generally speaking, higher temperature will cause the reaction rate to increase. And long residence time will allow for longer reaction time. Therefore we propose that a longer reactor with relatively high coolant fluid temperature, low inlet flow rate and low concentration of reactant will lead to a high outlet conversion. Countercurrent coolant fluid will have a better heat exchange performance thus a low conversion.

The simulation is performed in the COMSOL Multiphysics 5.5. Basically, our original reactor is 1 m long and 0.1m wide. It is assumed that the reactor is completely symmetrical in space, so 2D symmetric study is performed on this reactor. The reaction is a first order reaction. The rest of the information is listed in table 1. All parameters are varied around the initial value. The reactor radius changes from 0.01m to 0.2m and length of reactor changes from 0.5m to 2.5m. The outlet coolant temperature changes from the initial value 277K to 377K. The flow rate varies from half of the initial value, 0.05 mol/L, to twice of the initial value 2 mol/L. And the ratio of volume flow rate of water to propylene oxide and methane change from 1.5 to 3.5. A study on the reactant volumetric flow rate is done by changing its value from  $3\text{E-}5$  to  $1.18\text{E-}4$ . In each study, one parameter is varied while the rest are kept constant.

Table 1. Parameter of initial setting

Name	Expression	Description
E	75362[J/mol]	Activation energy
A	16.96e12[1/h]	Frequency factor
ke	0.559[W/m/K]	Thermal conductivity
Diff	1e-9[m <sup>2</sup> /s]	Diffusion coefficient
Uk	1300[W/m <sup>2</sup> /K]	Overall heat-transfer coefficient
dHrx	-84666[J/mol]	Heat of reaction
T0	312[K]	Inlet temperature of reactant
Ta0	277[K]	Inlet temperature of the coolant
v0	v_w0+v_po0+v_m0	Total flow rate
cA0	n_po0/v0	Propylene oxide concentration, inlet
cB0	n_w0/v0	Water concentration, inlet
cMe0	n_m0/v0	Methanol concentration, inlet
Cp0	(Cp_po*cA0+Cp_m*cMe0+Cp_w*cB0)/rho0	Heat capacity at inlet
rho0	(cA0*M_po+cB0*M_w+cMe0*M_m)	Density at inlet
Ra	0.1[m] Reactor	radius
L	1[m]	Reactor length

M_po	58.095[g/mol]	Molar weight, propylene oxide
M_m	32.042[g/mol]	Molar weight, methanol
M_w	18[g/mol]	Molar weight, water
rho_po_p	830[kg/m^3]	Density, propylene oxide
rho_m_p	791.3[kg/m^3]	Density, methanol
rho_w_p	1000[kg/m^3]	Density, water
Cp_po	146.54[J/mol/K]	Specific heat, po
Cp_m	81.095[J/mol/K]	Specific heat, m
Cp_w	75.36[J/mol/K]	Specific heat, w
Cp_pg	192.59[J/mol/K]	Specific heat, pg
v_ratio	3.5	Ratio water flow rate/(prop+me) flow rate
n_po0	0.1[mol/s]	Molar flow rate po in
n_m0	$v_{po0} \cdot \rho_{m\_p} / M_m$	Molar flow rate m in
v_po0	$n_{po0} \cdot M_{po} / \rho_{po\_p}$	Volumetric flow rate po in
v_m0	$v_{po0}$ Volumetric	flow rate m in
v_w0	$v_{ratio} \cdot (v_{po0} + v_{m0})$	Volumetric flow rate w in
n_w0	$\rho_{w\_p} \cdot v_{w0} / M_w$	Molar flow rate w in
Cpc	4180[J/(kg*K)]	Heat capacity, cooling fluid
mc	0.1[kg/s]	Mass flow rate, cooling fluid

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## Results

### Effect of Coolant Fluid Flow Direction

Figure 3 studies how the coolant flow direction affects the reaction, for concurrent and countercurrent models. These two figures show how temperature affects the conversion of reactants. The result shows that there is no obvious difference between the two flow directions. However, when the temperature is low, the concurrent flow case has a little higher outlet conversion than the countercurrent case.

Figure 4 continues studying the effect of flow direction. Here is the temperature distribution along the radial distance. The plots are nearly identical, which may explain why the outlet conversions for both flow directions are close to each other: more heat will be released as the reaction proceeds.

Figure 5 shows how changing flow rate affects the reaction in a tubular reactor. These two figures show the reactant concentration profiles of varying axial distances. By comparing the two plots, we can find that all reactants are almost consumed under the low flow rate case. Additionally, the concentration of reactants in low flow rate cases drops the most, indicating that in the second half of the reactor, more reaction takes place when the flow rate is low.

Figure 6 shows temperature distribution in the middle and outlet of the reactor. Unexpectedly, when the flow rate is lowered, the outlet temperature is higher rather than lower. The previous concentration profile can explain this: as the residence time increases, more reactions take place, thus resulting in a higher temperature.

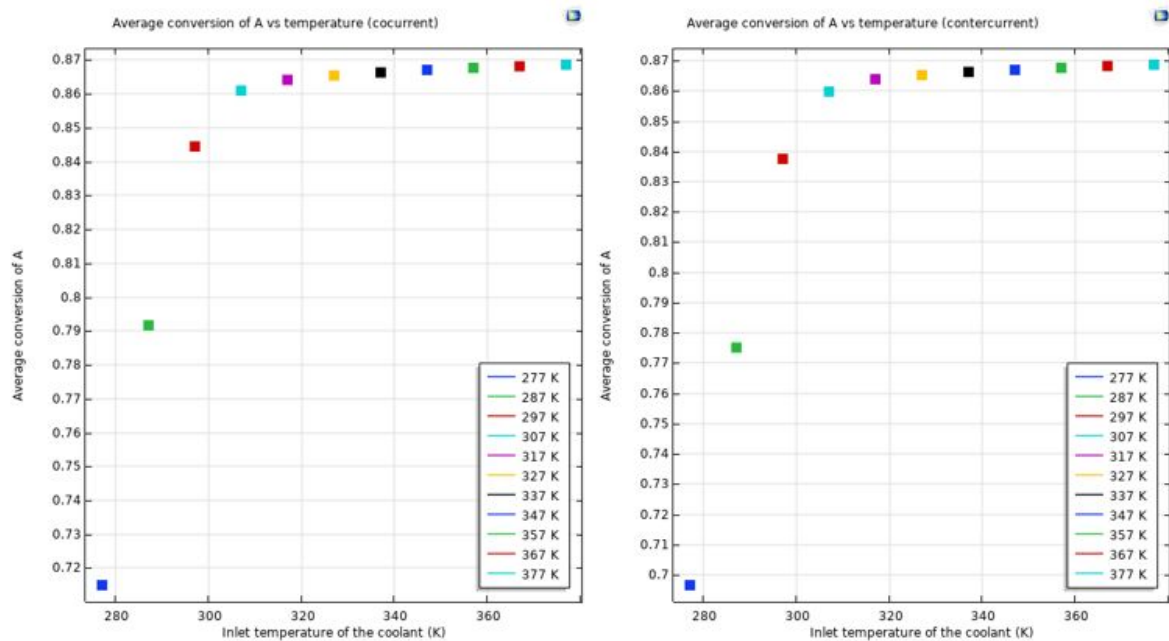


Figure 3

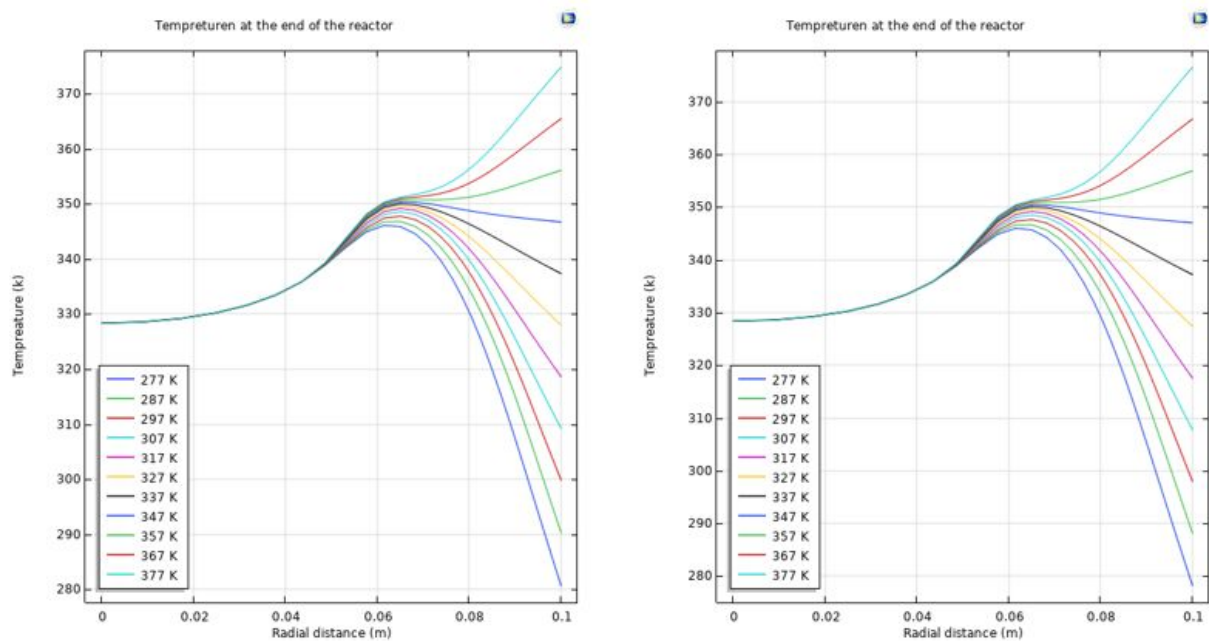


Figure 4

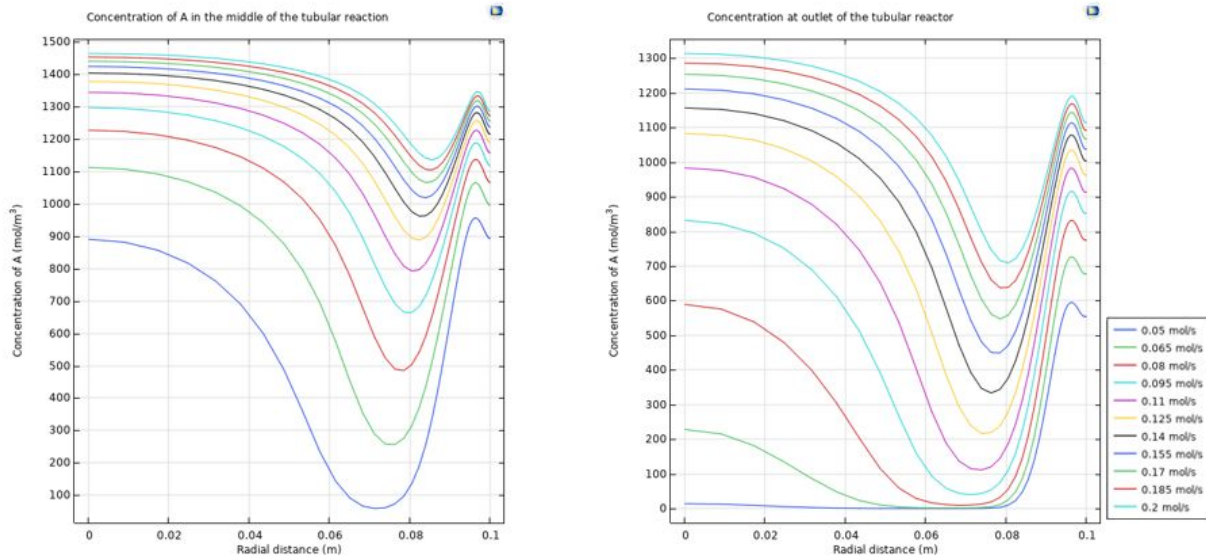


Figure 5

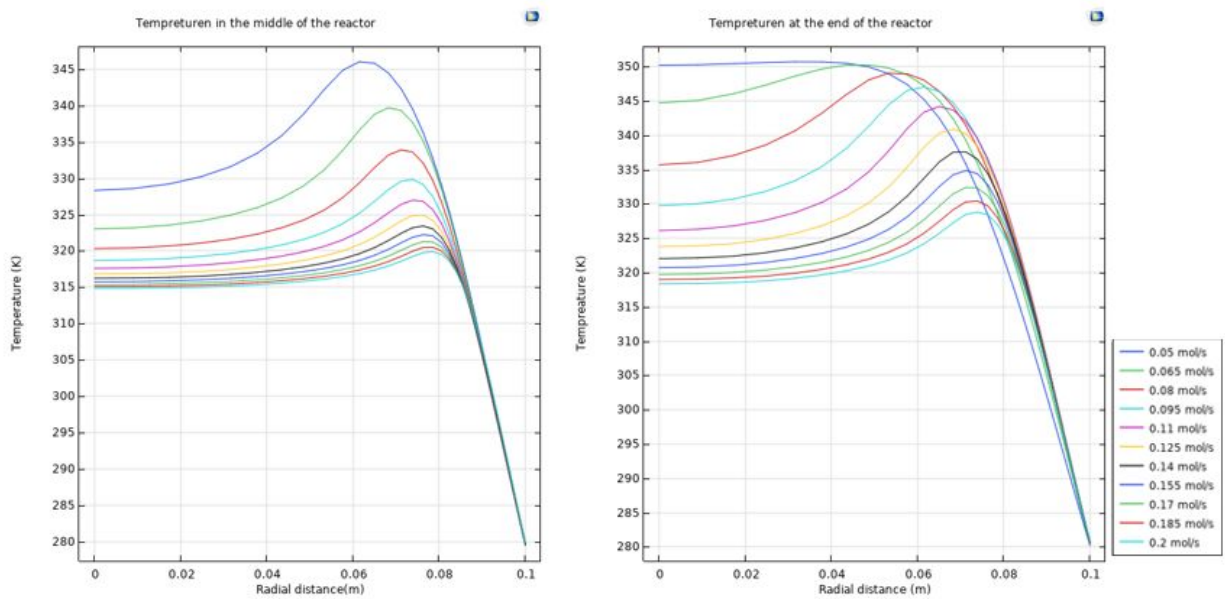


Figure 6

### Effect of Inlet Flow Rate

Volumetric Flow Rate is an important parameter in the design of a tubular reactor. In this section, the effect of varying flow rate is analysed on a second order reactor. The change in outlet conversion, temperature and concentration profiles at multiple locations are analyzed for

varying flow rate. The following has been studied in this section. The flow rate was varied from  $3.5\text{E-}5$  to  $1.18\text{E-}4 \text{ m}^3/\text{s}$ .

From Figure 7, it can be seen that as the flow rate is increased, the conversion decreases further. As the volume of the reactor has been treated constant for this part of the study, we can infer that the possible explanation for this behavior is the decrease in residence time with increasing flow rate. Due to decreased residence time, the outlet conversion begins to decrease.

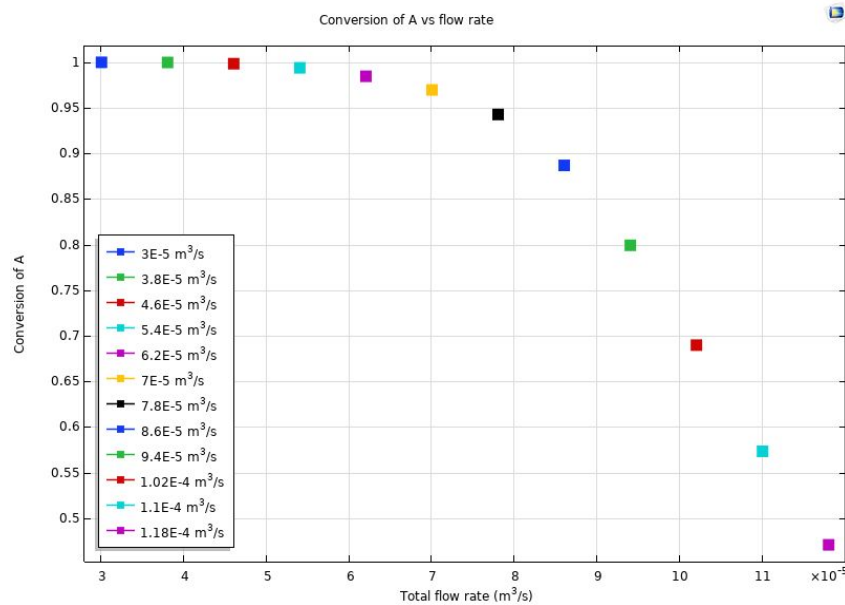


Figure 7

Based on the radial concentration profiles at the middle of the reactor in Figure 8, it can be observed that the concentration decreases continuously up to a point, then rising gradually and thereby having a slight dip near the walls.

The interpretation of this can be accounted to the assumption of a no-slip condition at the walls. Although there is a gradual decrease in the concentration as we proceed towards the end of the reactor, there is an increase near the walls.



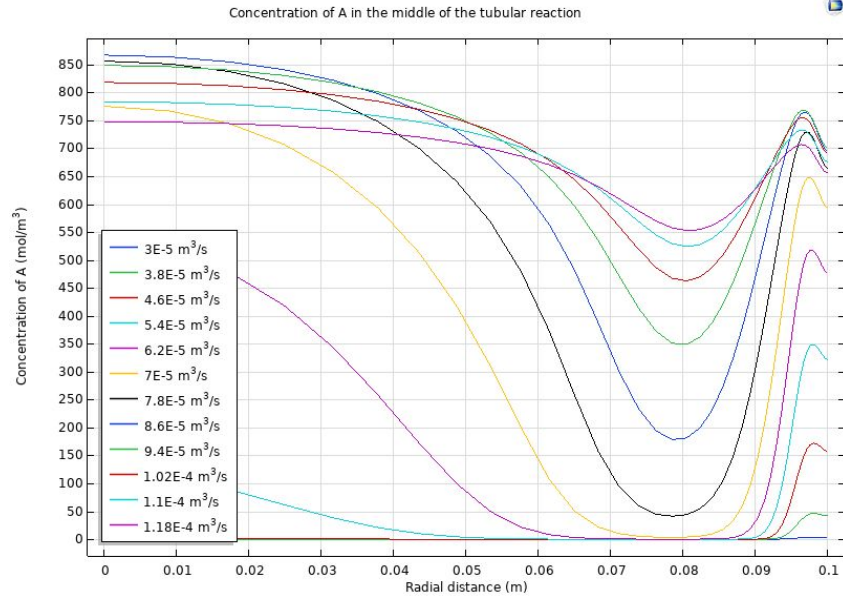


Figure 8

A similar behavior is observed at the end of the reactor, as shown in figure 9, with a subsequent increase in the concentration towards the end of the reactor. This can also be attributed to the no-slip condition which is assumed to be valid across the entire length of the reactor.

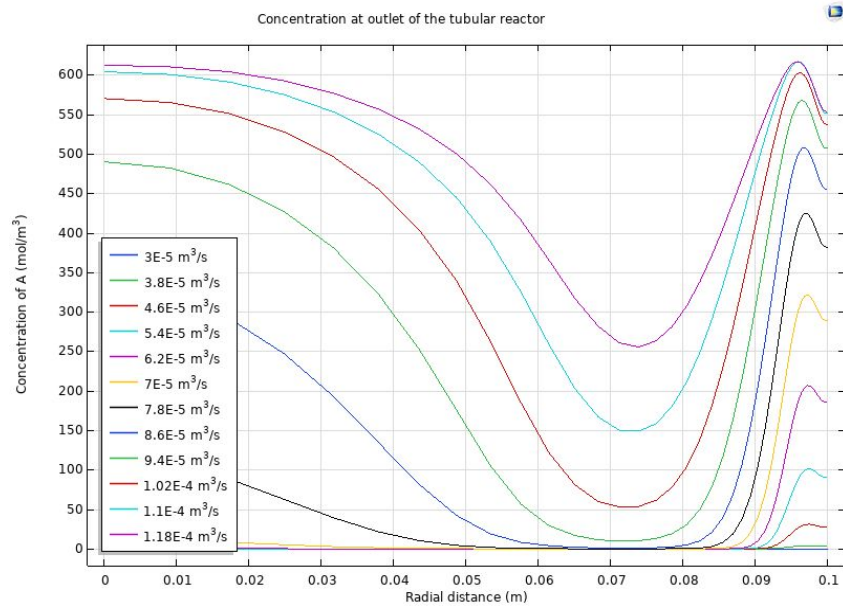


Figure 9

### Effect of Inlet Conditions on Temperature Profile

Based on the temperature profiles in the middle of the reactor, a general pattern of decreasing temperature across the radial direction is observed. From Figure 10, the temperature converges to the same value as we move towards the wall. This behavior is expected due to the no-slip condition. Also, based on our intuition, higher initial temperature gives rise to a higher overall temperature in the radial direction.

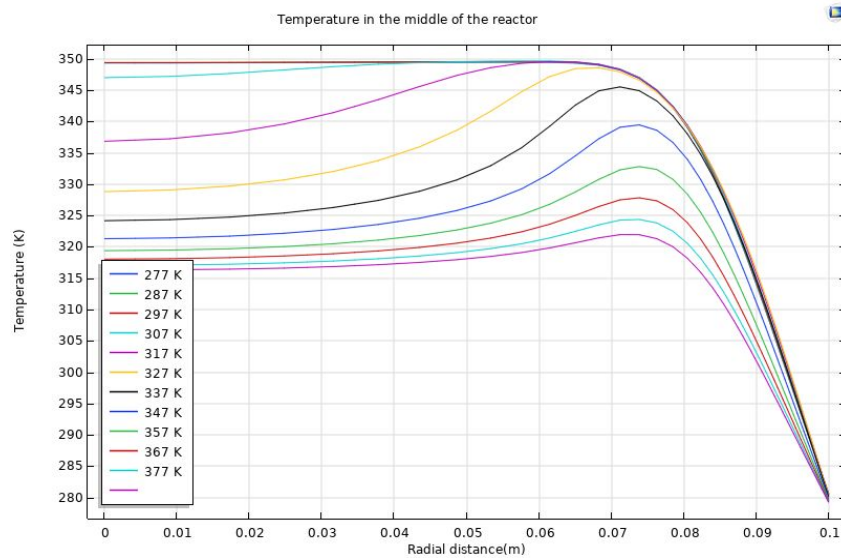


Figure 10

### Effect of Reactor Sizing

In this section, the effect of varying reactor radius and length is analyzed on an elementary reaction. The outlet conversion and temperature profiles are analyzed. The parametric sweep over the radius is done by keeping the length constant at 1m and varying the radius from 0.05m to 0.2m. Likewise, the parametric sweep over the length is done by keeping the radius constant at 0.1m while changing the length from 0.05m to 2.5m.

As the radius increases from 0.05m to 0.2m, as shown in figure 11, the outlet conversion of the species A at the center of the tubular reactor also increases. As the radius is approaching 0.15m, the conversion at the center converges to 1. After the radius has reached 0.2m, which is not shown in the figure below, the conversion at both the center and the wall of the reactor converges to 1. The generated convergence profile presents a meaningful scale for a tubular reactor design when a specific convergence at the outlet is required.

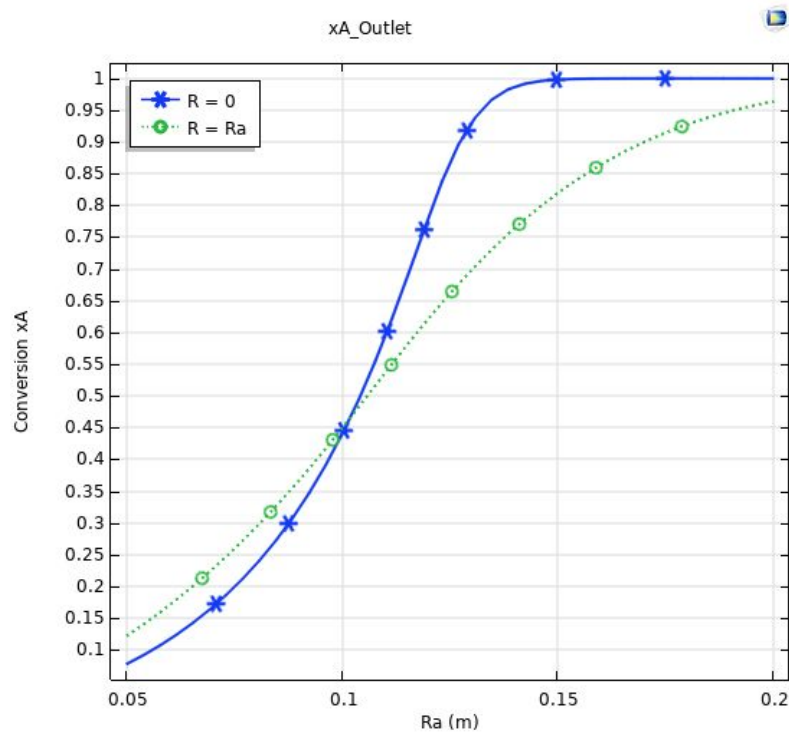


Figure 11

The temperature profile, as shown in figure 12, corresponds to the conversion profile. The outlet temperature at the center of the reactor increases as its radius increases. This is due to the fact that the outlet conversion increases with the radius and the reaction being exothermic.

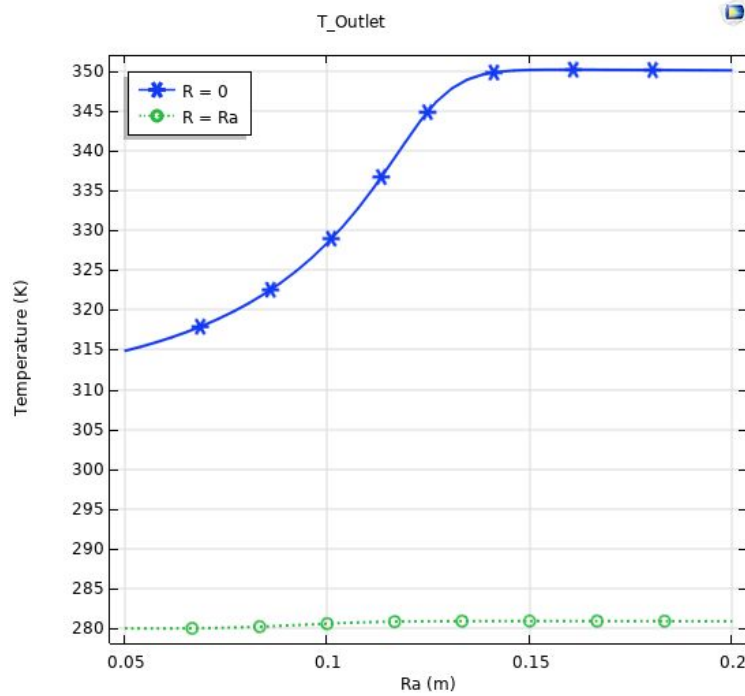


Figure 12

Figure 13 shows the outlet conversion profile at both the center and wall of the tubular reactor as the length changes from 0.05m to 2.5m, while the radius is kept constant at 0.1m. As the reaction proceeds along the length, the conversion at the outlet increases. The conversion across the intersection of the tubular reaction exceeds 0.8 as the length reaches 1m. The conversion profiles generated by parametric sweep over both radius and length give valid guidance on the reactor design for the specific reaction studied. For example, for this propylene glycol production with required outlet conversion of 0.8, an optimal reactor sizing might suggest a radius at 0.1m and length at 1m.

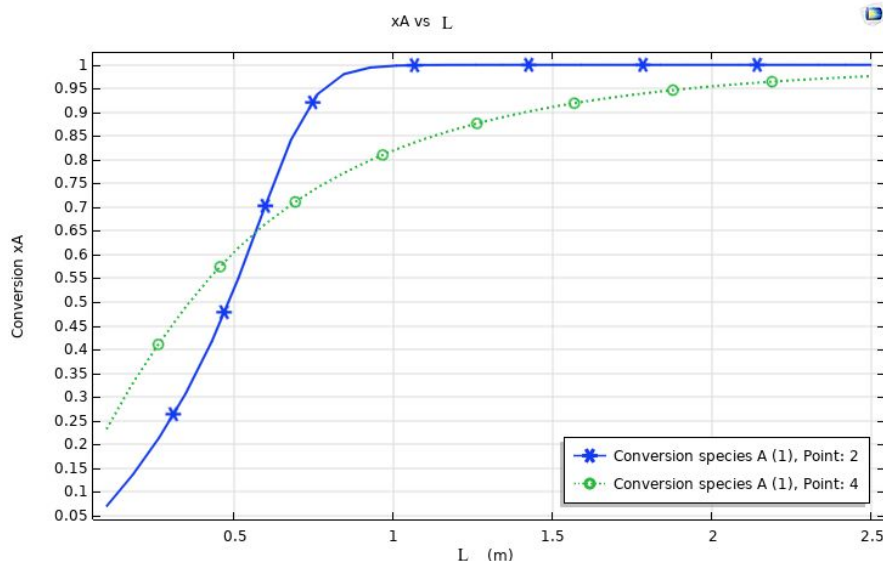


Figure 13

### Effect of Inlet Concentration

In this section, we analyze how inlet concentration affects concentration and temperature profiles in the reactor. The inlet concentration of the model is set up as the ratio between water and propylene oxide. This mechanism allows us to perform an auxiliary sweep on the volumetric ratio ranging from 3.5 to 1.5 to study the effect of different concentrations. One thing to note is that higher water to propylene ratio is equivalent to lower concentration.

Figures 14 and 15 show the temperature and concentration profiles for the first order reaction. As expected, higher concentration results in higher reaction rate, which can be observed both in the conversion of propylene oxide and temperature of the mixture. At the highest concentration, 1.5 water/propylene oxide, the reactant fully reacted at 0.4 m of the reactor, while only 45% conversion was achieved at the end of the reactor. These figures give us a reference to the reactor design. If we would like to fully utilize the whole reactor, we should set up inlet concentration at around a volumetric ratio of 2.5.

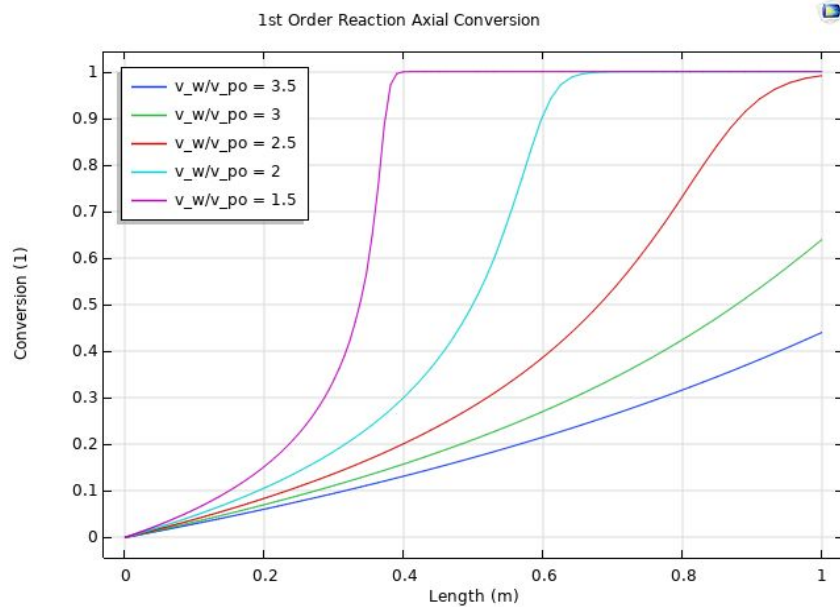


Figure 14

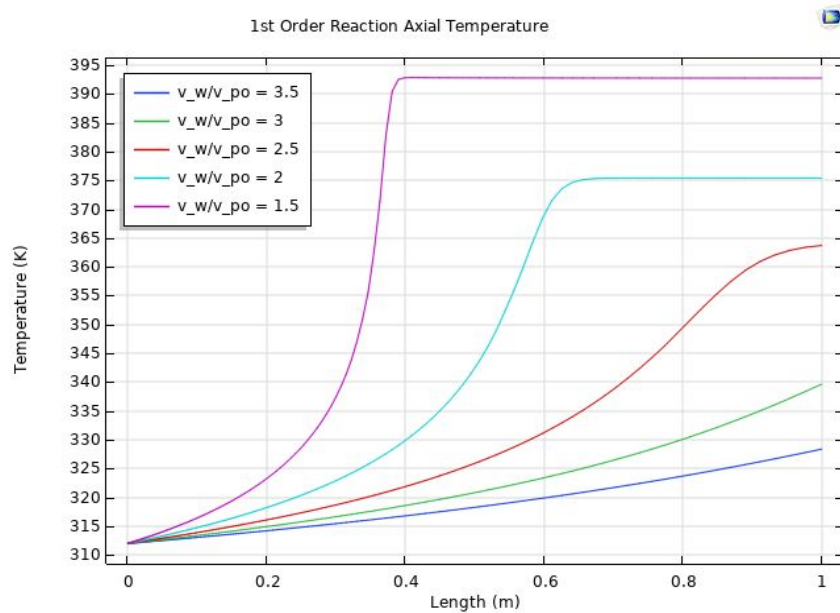


Figure 15

Figures 16 and 17 show the temperature and concentration profiles for the second order reaction. From these figures, we can see that the reaction also follows the rule that higher concentrations result in higher reaction rates. However, the reaction rates are much higher compared to that of

the first order mechanism. All reactants are fully converted to products at around 0.3 m. To obtain an efficient design of the reactor, the length can be shortened to reduce cost.

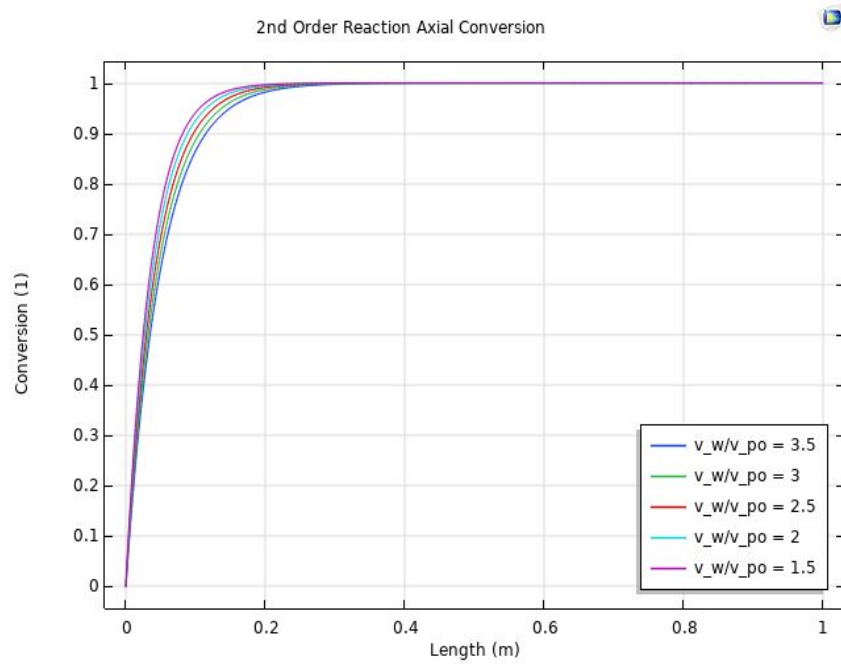


Figure 16

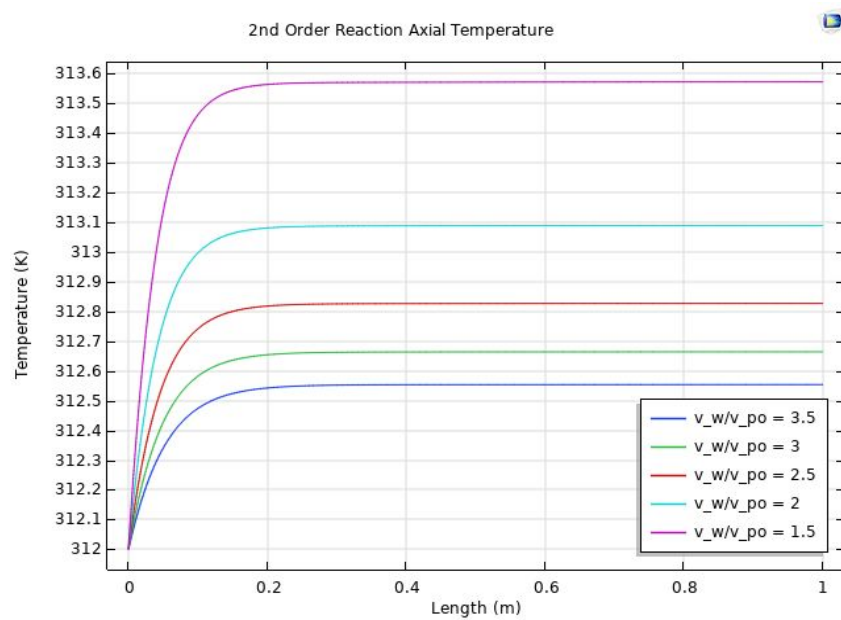


Figure 17

### Mesh Independence Study

To justify our mesh selection, we study mesh independence. A probe is set up at  $r = 0.02$  m and  $z = 0.5$  m; and two parametric sweeps for vertical and horizontal numbers of elements are performed. The probe is shown in Figure 18, and concentration under different meshes are shown in Figure 19 and 20. From the results, our choice of 50 elements vertically and 200 elements horizontally is quite reasonable.

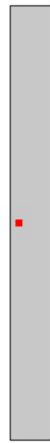


Figure 18

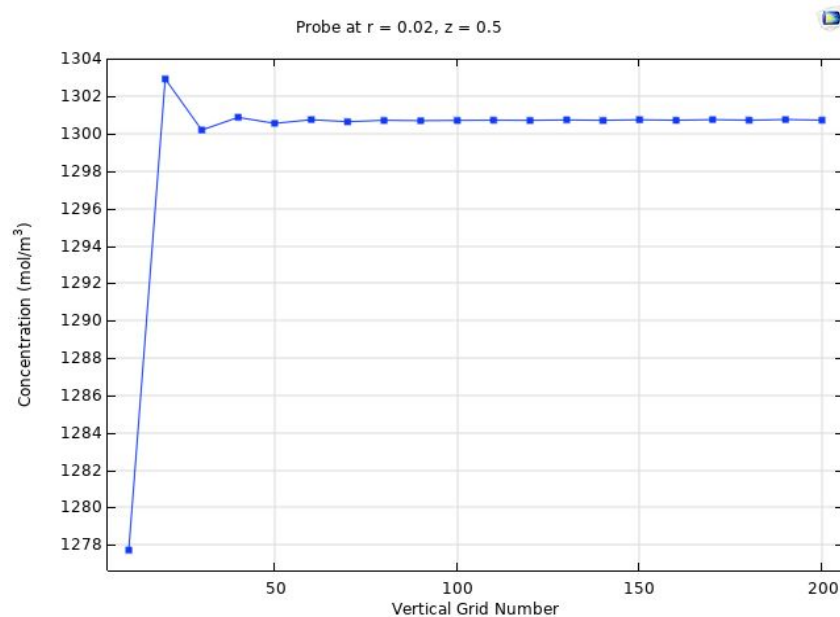


Figure 19



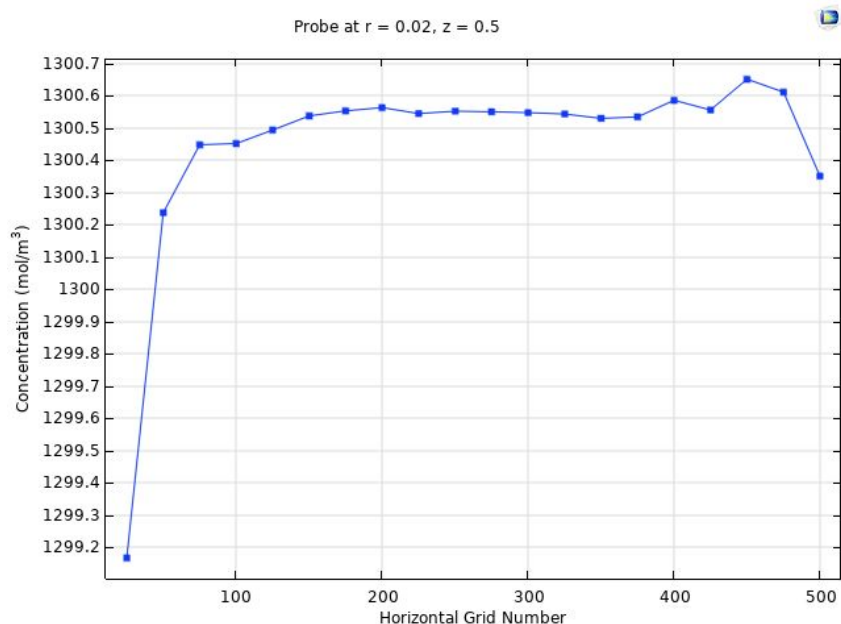


Figure 20

## Conclusion

In conclusion, we find that a longer reactor should be implemented if a higher conversion is desired. Also, decreasing the flow rate, concentration, and ratio of propylene oxide to water can help increase the conversion at the outlet of the reactor. However, these three strategies will increase the temperature significantly. Therefore, a better heat exchange device is required to keep the reactor at an appropriate temperature. Increasing the temperature of the coolant fluid can also increase the conversion. However, the flow direction of coolant fluid has no effect on the conversion. Moreover, the second order reaction assumed in this project will take place much faster than the first order reaction. More research can be conducted in the future on more complicated reaction mechanisms to determine the feasibility of this simple tubular reactor.

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