ADAPTIVE NEURO-FUZZY INFERENCE SYSTEM (ANFIS) –
BASED MODEL PREDICTIVE CONTROL (MPC)
FOR CARBON DIOXIDE REFORMING OF METHANE (CDRM)
IN A PLUG FLOW TUBULAR REACTOR FOR HYDROGEN
PRODUCTION

A Thesis
Submitted to the Faculty of Graduate Studies and Research
In Partial Fulfillment of the Requirements for the Degree of
Master of Applied Science
in
Industrial Systems Engineering
University of Regina

by

Ememobong Ita Essien
Regina, Saskatchewan
January, 2013

Copyright 2012: Ememobong Ita Essien
Ememobong Ita Essien, candidate for the degree of Master of Applied Science in Industrial Systems Engineering, has presented a thesis titled, *Adaptive Neuro-Fuzzy Inference Systems (ANFIS) - Based Model Predictive Control (MPC) for Carbon Dioxide Reforming of Methane (CDRM) in a Plug Flow Tubular Reactor for Hydrogen Production*, in an oral examination held on November 28, 2012. The following committee members have found the thesis acceptable in form and content, and that the candidate demonstrated satisfactory knowledge of the subject material.

External Examiner: Dr. Shahid Azam, Environmental Systems Engineering

Co-Supervisor: Dr. Hussameldin Ibrahim, Process Systems Engineering

Co-Supervisor: Dr. Mehran Mehrandezh, Industrial Systems Engineering

Co-Supervisor: Dr. Raphael Idem, Industrial Systems Engineering

Committee Member: Dr. Ezeddin Shirif, Petroleum Systems Engineering

Committee Member: Dr. David deMontigny, Process Systems Engineering

Chair of Defense: Dr. Haithem Zourrig, Faculty of Business Administration

*Not present at defense*
ABSTRACT

The current sources of our energy supply are plagued with many problems, and the impact on the climate is of grave concern. To preserve and sustain our environment, a non-polluting and renewable energy source is required. Hydrogen (H₂), when extracted from one of its many sources during carbon dioxide (CO₂) capture, is considered a non-polluting, efficient and environmentally sustainable energy source.

In this research work, the control of a pilot-scale reformer for the production of hydrogen was studied. Hydrogen was produced through the carbon dioxide reforming of methane (CDRM). This process was used to convert methane (CH₄) and carbon dioxide into hydrogen. A high methane conversion was maintained by controlling the temperature in the reformer at the thermodynamically desired level.

The control strategy applied to this process was the model predictive control (MPC) based on an adaptive neuro-fuzzy inference system (ANFIS) model. MPC has, among other advantages, the ability to predict the response of the system over a given prediction horizon. Experimental results showed that the ANFIS model was able to accurately replicate the response of the process to changes in temperature. Based on the ANFIS model, an MPC strategy was formulated for the process.
ACKNOWLEDGEMENTS

My sincere gratitude goes to my parents and my siblings for being my backbone throughout the duration of my program. Thank you immensely for all the words of encouragement that kept me going when I needed it the most.

The role my supervisors, Dr Raphael Idem, Dr Hussameldin Ibrahim and Dr Mehran Mehrandezh, played in ensuring the successful completion of this research work cannot be overemphasized. I greatly appreciate your guidance, assistance and constructive feedback for the length of the program. I would not have made it this far without your support, and for that, I say thank you.

In the same vein I am grateful to the Faculty of Graduate Studies and Research of the University of Regina for the opportunity to carry out this research work and also for providing financial support.

I also want to express my gratitude to Elendus, Onasanyas, Omoyefas, and Tettevis for their prayers and moral support, as well as the contributions of my friends, colleagues and everyone who made my program pleasurable.

I am indebted to Delasi Tettevi, whose contributions to this work are invaluable. I want to say a big thank you for all your sacrifices.

Most importantly, my sincere appreciation goes to the Almighty God who endowed me with the ability to put forward the ideas contained in this thesis. I am most grateful to God for the grace and strength he gave to me to carry out this research and to see it to fruition.
DEDICATION

To my parents for their love, support and encouragement
# TABLE OF CONTENT

ABSTRACT..............................................................................................................ii
ACKNOWLEDGEMENTS.........................................................................................iii
DEDICATION...........................................................................................................iv
TABLE OF CONTENTS............................................................................................v
LIST OF TABLES.......................................................................................................ix
LIST OF FIGURES.....................................................................................................x
NOMENCLATURE....................................................................................................xiv

CHAPTER 1- INTRODUCTION.....................................................................................1
1.1 Energy Systems....................................................................................................1
1.2 Control Theory.....................................................................................................3
1.3 Model-based Control Strategies..........................................................................5
1.4 Optimal Control Theory.......................................................................................6

1.4.1 Introduction to Model Predictive Control (MPC)............................................7
1.5 Statement of the Problem.....................................................................................8
1.6 Scope and Objective of Research Work..............................................................9
1.7 Organization of Thesis........................................................................................10

CHAPTER 2- LITERATURE REVIEW........................................................................11
2.1 Hydrogen Production..........................................................................................11
2.2 Hydrogen Production from Conventional Sources...........................................14

2.2.1 Natural Gas Steam Reforming......................................................................14
2.2.2 Partial Oxidation of Hydrocarbons..............................................................15
2.2.3 Coal Gasification..........................................................................................16
2.3 Hydrogen Production from Biomass and Renewable Sources..............................17
2.4 Carbon Dioxide Reforming of Methane...............................................................18
2.5 Tubular Reactors and Temperature Control.........................................................19
2.6 Applications of Model Predictive Control (MPC) Strategy.................................21
   2.6.1 Application of MPC in Robotics.................................................................22
   2.6.2 Application of MPC in Medicine.................................................................23
   2.6.3 Application of MPC in Process Systems......................................................24
2.7 Models in the MPC Strategy..................................................................................26
   2.7.1 MPC Strategy Based on Linearized Models of the Process.........................27
   2.7.2 Nonlinear MPC Strategies.............................................................................27
   2.7.3 Neural Network Based MPC..........................................................................28
   2.7.4 Soft Computing and Control.........................................................................29
       2.7.4.1 Adaptive Neuro-Fuzzy Inference System (ANFIS) Modeling and Identification.................................................................29
2.8 MPC Strategy Based on ANFIS Models...............................................................32

CHAPTER 3- METHODOLOGY.....................................................................................34
3.1 Model Development..............................................................................................34
3.2 Chemical Reactor Modeling................................................................................36
   3.2.1 The Plug Flow Reactor...................................................................................40
   3.2.2 The Packed Bed Tubular Reactor...................................................................41
       3.2.2.1 The One Dimensional Reactor Model..................................................42
       3.2.2.2 The One Dimensional Reactor Model with Axial Mixing.................43
       3.2.2.3 The Two Dimensional Pseudo-Homogeneous Models......................45
3.3 The Simulation Model..........................................................................................46
3.4 Model Predictive Control (MPC) Theory............................................................48
3.4.1 MPC Strategy

3.5 Nonlinear Predictive Control

3.6 Nonlinear System Identification

3.6.1 Nonlinear System Identification using a Fuzzy Model

3.6.2 Nonlinear System Identification using a Neuro-Fuzzy Model

3.6.2.1 Adaptive Neuro-Fuzzy Inference System (ANFIS) Architecture

3.6.2.2 Hybrid Learning Algorithm

3.7 COMSOL Multiphysics

3.7.1 Chemical Reaction Engineering Module

3.7.2 Transport of Diluted Species Interface

3.7.3 Heat Transfer Module

3.7.4 Heat Transfer in Fluids Interface

CHAPTER 4- ANALYSIS AND RESULTS

4.1 Controller Design Strategy

4.2 Phase 1 - Steady State Simulation of the Plug Flow Reactor in Comsol Multiphysic

4.3 Phase 2 - Transient Response of the Plug Flow Reactor

4.4 Phase 2 - Response of Reactor to Step Change in External Temperature

4.5 Phase 2 - The Flow Rate Term ($V_w$)

4.6 Phase 3 - Data for ANFIS Training

4.7 Phase 3 - ANFIS Model and Training

4.8 Phase 4 – Control Strategy Implementation
CHAPTER 5- LIMITATIONS, CONCLUSIONS AND RECOMMENDATIONS.118

5.1 Limitations.................................................................118
5.2 Conclusions............................................................119
5.3 Recommendations for Future Studies.................................120

REFERENCES.................................................................121
LIST OF TABLES

Table 4.1 Operating conditions and parameters used in the simulation…………….69

Table 4.2 Selection of inputs for the ANFIS model……………………………….99

Table 4.3 Number of rules for the models considered…………………………101
LISTS OF FIGURES

Figure 1.1  Open loop control system…………………………………………………………3

Figure 1.2  Closed loop control system………………………………………………………4

Figure 1.3  Schematic diagram of the experimental setup for the $CO_2$ dry reforming of methane using packed bed tubular reactor (Akpan et al., 2007)………9

Figure 2.1 Alternative methods of hydrogen production (Balat, 2009)………………12

Figure 2.2 Feedstock used in the current global production of hydrogen (Kothari et al., 2006)………………………………………………………………………………13

Figure 2.3 Procedure for the design of an MPC strategy………………………………33

Figure 3.1 Interaction between the system and its environment (Roffel and Betlem, 2006)………………………………………………………………………………38

Figure 3.2 MPC Strategy (Espinosa et al., 2005)………………………………………51

Figure 3.3 A two-input first-order Sugeno fuzzy model with two rules (Jang et al., 1997)…………………………………………………………………………………59

Figure 3.4 Equivalent ANFIS architecture (Jang et al., 1997)…………………………59

Figure 3.5 Piecewise linear approximation of consequent MFs in Tsukamoto ANFIS (Jang et al., 1997)…………………………………………………..……63
Figure 4.1 Surface plot of concentration…………………………………………70
Figure 4.2 Surface plot of conversion…………………………………………72
Figure 4.3 Surface plot of temperature…………………………………………73
Figure 4.4 Methane conversion profile in the axial direction………………..74
Figure 4.5 Transient response of the reactor (Conversion profile)………………75
Figure 4.6 Transient response of the reactor measured at 3 points at the reactor outlet………………………………………………………………76
Figure 4.7 Transient response of the reactor showing how long it takes to get to steady state…………………………………………………………77
Figure 4.8 Transient response of the reactor (Concentration profile)……………78
Figure 4.9 Transient response of the reactor (Temperature profile)……………79
Figure 4.10 Step change in external temperature .................................80
Figure 4.11 Response of reactor to step change in external temperature (conversion profile)…………………………………………………………81
Figure 4.12 Response of reactor to step change in external temperature (concentration profile)…………………………………………………82
Figure 4.13 Response of reactor to step change in external temperature (temperature profile)……………………………………………………83
Figure 4.14 Conversion for various flow rates

Figure 4.15 Step change in flow rate in outer jacket

Figure 4.16 Response of reactor to step change in flow rate (conversion profile)

Figure 4.17 Response of reactor to step change in flow rate (concentration profile)

Figure 4.18 Response of reactor to step change in flow rate (temperature profile)

Figure 4.19 Conversion profile at 1073 K

Figure 4.20 Random change in flow rate

Figure 4.21 Reactor response to random change in flow rate (conversion profile)

Figure 4.22 Reactor response to random change in flow rate (concentration profile)

Figure 4.23 Reactor response to random change in flow rate (temperature profile)

Figure 4.24 Validation results for the reactor

Figure 4.25 Membership function of the neuro-fuzzy model of the reactor before training

Figure 4.26 Membership function of the neuro-fuzzy model of the reactor after training
Figure 4.27 Error plots for the training process ..............................107
Figure 4.28 Output response to setpoint change .............................111
Figure 4.29 Change in control signal due to setpoint change ..............112
Figure 4.30 Change in deltau due to setpoint change .......................113
Figure 4.31 Response of system to “stair” reference ..........................114
Figure 4.32 Applied perturbations ..............................................115
Figure 4.33 Response of system to applied perturbations ....................116
**NOMENCLATURE**

*Notations*

<table>
<thead>
<tr>
<th>Symbol</th>
<th>Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>$A_i$</td>
<td>linguistic label</td>
</tr>
<tr>
<td>$B_i$</td>
<td>linguistic label</td>
</tr>
<tr>
<td>$C$ or $C_A$ or $C_i$</td>
<td>feed concentration of species (mol/m$^3$)</td>
</tr>
<tr>
<td>$C_0$ or $C_{A_0}$ or $C_{i_0}$</td>
<td>initial concentration of species $i$ (mol/m$^3$)</td>
</tr>
<tr>
<td>$c_p$</td>
<td>heat capacity (kJ/kg.K)</td>
</tr>
<tr>
<td>$c_{pg}$</td>
<td>heat capacity of the gas mixture (kJ/kg.K)</td>
</tr>
<tr>
<td>$c_{pm}$</td>
<td>heat capacity of the methane (kJ/kg.K)</td>
</tr>
<tr>
<td>$c_v$</td>
<td>heat capacity at constant volume</td>
</tr>
<tr>
<td>$(CH_4)_{in}$</td>
<td>concentration of the methane feed (mol/m$^3$)</td>
</tr>
<tr>
<td>$(CH_4)_{out}$</td>
<td>concentration of methane measured at the reactor outlet (mol/m$^3$)</td>
</tr>
<tr>
<td>$d_i$ or $D_i$</td>
<td>internal diameter of reactor (m)</td>
</tr>
<tr>
<td>$D_{ea}$</td>
<td>effective axial diffusivity (m$^2$/h)</td>
</tr>
<tr>
<td>$D_{er}$</td>
<td>effective radial diffusivity (m$^2$/h)</td>
</tr>
<tr>
<td>$D_{eff}$</td>
<td>effective diffusivity (m$^2$/h)</td>
</tr>
<tr>
<td>$E$</td>
<td>activation energy (kJ/kmol)</td>
</tr>
<tr>
<td>$f(.,.)$</td>
<td>dynamic model of the plant</td>
</tr>
<tr>
<td>$F_i$</td>
<td>molar flow rate of the chemical species (kmol/h)</td>
</tr>
<tr>
<td>$f_j$</td>
<td>consequence of the $j$th fuzzy rule</td>
</tr>
</tbody>
</table>
\( g(\cdot, \cdot) \) \hspace{1cm} \text{dynamic model of the plant}

\( g_t \) \hspace{1cm} \text{step response coefficient of the plant}

\( \Delta H \) \hspace{1cm} \text{heat of reaction (kJ/kmol)}

\( J \) \hspace{1cm} \text{objective function}

\( k \) \hspace{1cm} \text{fluid thermal conductivity}

\( k_0 \) \hspace{1cm} \text{collision coefficient}

\( K_A \) \hspace{1cm} \text{adsorption constant for component A}

\( K_P \) \hspace{1cm} \text{thermodynamic equilibrium constant}

\( L \) \hspace{1cm} \text{length of catalyst bed (m)}

\( n \) \hspace{1cm} \text{reaction order}

\( N \) \hspace{1cm} \text{data array for training}

\( N_c \) \hspace{1cm} \text{control horizon.}

\( N_p \) \hspace{1cm} \text{prediction horizon.}

\( O_{ij} \) \hspace{1cm} \text{output of the} j \text{th node in the} i \text{th layer}

\( p_j \) \hspace{1cm} \text{consequent parameter of the} j \text{th fuzzy rule}

\( P_{tot} \) \hspace{1cm} \text{total pressure within the reactor (kPa)}

\( q_j \) \hspace{1cm} \text{consequent parameter of the} j \text{th fuzzy rule}

\( \dot{q} \) \hspace{1cm} \text{total heat flux into the system (W/m}^2\text{)}

\( Q \) \hspace{1cm} \text{heat source (W/m}^3\text{)}

\( Q_{ctl} \) \hspace{1cm} \text{positive definite matrix}

\( r \) \hspace{1cm} \text{radial direction of the reactor}

\( r_A \text{ or } r_i \) \hspace{1cm} \text{rate of reaction}
\( r_j \) consequent parameter of the \( j \)th fuzzy rule

\( R \) gas constant (kJ/kmol.K)

\( R_{ctt} \) positive semi-definite matrix

\( R_i \) internal radius of reactor (m)

\( S_{ctt} \) positive semi-definite matrix

\( T_0 \) feed temperature (K)

\( T(r, z) \) or \( T \) reactor temperature (K)

\( T_w \) temperature of the heating fluid (K)

\( u_z \) superficial velocity of the fluid (km/h)

\( u(t) \) or \( u(k) \) control input

\( \hat{u}(k) \) estimated input

\( du \) step size

\( \delta u \) first element of \( \Delta U \)

\( U \) or \( U_{tw} \) overall heat transfer coefficient (kJ/m\(^2\).h.K)

\( \Delta U \) change in control signal

\( v_i \) stoichiometric coefficient of component \( i \)

\( V \) total volumetric flow rate of the feed (m\(^3\)/s)

\( V_w \) total volumetric flow rate of the heating fluid (m\(^3\)/s)

\( w(t) \) or \( w(k) \) reference setpoint

\( w_i \) firing strength of the fuzzy rules

\( \bar{w}_i \) normalized firing strength

\( x(k) \) system state
\( X(k) \) conversion

\( y(t) \) or \( y(k) \) system output

\( \hat{y}(t) \) estimated output

\( z \) axial direction of the reactor

\textit{Symbols}

\( \alpha_w \) heat transfer coefficient near the reactor wall

\( y \) ratio of specific heat

\( \theta \) vector of parameters to be chosen

\( \mu_i \) membership function

\( \lambda_{ea} \) effective axial thermal conductivity (kJ/m.s.K)

\( \lambda_{er} \) effective radial thermal conductivity (kJ/m.s.K)

\( \lambda_{eff} \) effective thermal conductivity (kJ/m.s.K)

\( \rho_m \) density of the fluid in the heat exchanger (kg/m\(^3\))

\( \rho_B \) bulk density (kg/m\(^3\))

\( \rho_g \) gas density (kg/m\(^3\))

\( \varphi \) regression vector

\( \nabla \) gradient function
1. INTRODUCTION

1.1 Energy Systems

The problems with the existing energy systems in the modern world are perceived as many, reaching from security of supply to climate change issues (Balat, 2009). Presently our over-dependence on fossil fuels is eroding our environment at a slow but steady rate (Jin and Jin, 2010). Moreover the current energy sources are non-renewable and depleting at an alarming rate. This trend has been encouraged by the continuous growth in global population and economic development, thereby leading to an increase in the concentration of carbon dioxide, methane and nitrous oxide in the environment.

A major challenge facing this generation has been to provide more energy while limiting greenhouse gas (GHG) emissions. Unfortunately these GHGs are usually the by-product of the combustion of natural gas (NG), coal, and oil, which are fossil fuels. These are used for heating, electricity production, transportation, and industrial purposes, which are vital for the existence of man-kind (Padin et al., 2000; Abbas and Wan Daud, 2010).

For the safety of our environment, there is an urgent need to develop non-polluting and renewable energy sources. This is evident in both political statements and demonstration projects all over the world. One version of the vision for a sustainable energy system that has been able to unite economic growth and environmental concerns is the vision of the hydrogen economy (Saxe and Alvfors, 2007).
Hydrogen is considered a non-polluting, efficient, inexhaustible energy carrier alternative to fossil fuels for the future (Navarro et al., 2007). It is a promising renewable fuel for transportation and domestic applications. Currently, research work is being done on the application of hydrogen in combustion engines and fuel-cell electric vehicles (U.S. Department of Energy, 2012). Hence, it is safe to say that in both the near term and long term, hydrogen demand is expected to increase significantly. The amount of energy produced during hydrogen combustion is higher than that by any other fuel on a mass basis. It is estimated to be 2.4, 2.8, or 4 times higher than that of methane, gasoline, or coal, respectively. Currently the annual production of hydrogen is about 0.1 gigaton, 98% of that is from the reforming of fossil fuels (Abbas and Wan Daud, 2010).

Contemporary researchers, Akpan et al. (2007), engaged in research work that sought to convert two of the aforementioned greenhouse gases: carbon dioxide and methane ($CO_2$ and $CH_4$ respectively) to hydrogen. They conducted kinetic, experimental, modeling, and simulation studies of carbon dioxide reforming of methane (CDRM). This study was carried out in a catalytic packed bed tubular reactor using a $Ni/CeO_2-ZrO_2$ catalyst. The current research work is based primarily on this initial work; however, this work focuses on the design of an appropriate control strategy to maintain the desired conditions within the reactor to ensure an active and stable methane conversion reaction.
1.2 Control Theory

Control theory deals with the various disciplines and methods that lead to an automatic decision process. The outcome of this decision process usually enhances the performance of the system (Bars et al., 2006). Industrial control systems are usually exposed to noisy and polluted environment, and they are subjected to unpredictable disturbances. It is the function of the control system to suppress the effects of these external disturbances by changing the overall system characteristics, in order to compensate for these disturbances (Shaw, 1998).

The control strategy could either be an open-loop or closed-loop control. In open-loop control, no feedback mechanism is used in enforcing the desired output set point. This control strategy relies fully on the current state and the model of the system to calculate its control inputs.

![Figure 1.1 Open loop control system](image)

Figure 1.1 Open loop control system

In Figure 1.1 above, a process has been designed together with a controller to achieve the desired response. The boxes represent the functional relationships that exist between the signals, indicated by the arrows. The design above is an open loop system, in that there is no monitoring of the actual system response.
This can be illustrated with the example of a car being driven from point A to point B. The driver and the steering mechanism serve as the controller. An open loop system can be achieved if the driver is blind-folded. As long as it is a straight road from point A to point B, and the driver maintains his trajectory, accidents would be averted. In essence an open loop system functions properly only if all design specifications are maintained: any disturbance will produce inaccurate results.

On the other hand a closed loop control strategy is one that the system inputs are determined, at least in part, by the system outputs. Thereby making the system inputs a function of the system outputs, and vice versa. This is shown in the diagram below:

![Diagram of a closed loop control system](image)

Figure 1.2 Closed loop control system

Figure 1.2 shows the general structure of a closed loop system. The actual system response is measured and then compared to the desired output. The feedback monitors the performance of the system and then uses this information for the proper control of
the system. The advantage of introducing a feedback mechanism is that it allows the system to respond to any disturbances that act upon it from the environment.

For the closed loop system, using the same example as above, the driver is the feedback for the system when his eyes are open. He monitors the car to ensure it is traveling in the correct direction, and determines if the actual response is identical to the desired response. The driver, together with the steering mechanism of the car, also serves as the controller of the system, thereby introducing any changes in the direction of travel so that the desired response is achieved. If the road curves in a certain direction, this information is relayed to the driver through his vision and the needed modification in the direction of travel is introduced. A closed loop system constantly monitors the output of the process under control; any disturbance that affects the system’s response will result in an appropriate modification of the system’s operation so that the desired output will be obtained.

1.3 Model-Based Control Strategies

Control design and implementation uses methods, which are either model-free or model based. Models are very important in our everyday lives, and are employed in all aspects of human activities. Most of the models used are qualitative in nature and not explicitly formulated even though they exist in our everyday world and hence they cannot be reproduced, verified or validated. In complex industrial systems however, it is imperative that one fully understands the process behavior of the system model. These models are therefore, expressed in mathematical terms. Control designs based on this concept are termed Model-Based Control (MBC) and most modern control
design techniques defer to this conceptualization. Models used in control system designs could either be linear or non-linear.

1.4 Optimal Control Theory

A major challenge facing control engineers is that of reducing costs while maintaining or improving product quality. In tackling this challenge, the safety and integrity of the system must not be compromised. As systems become more complex, it is important to ensure that the implemented systems are reliable (Bars et al., 2006).

Another consideration for control engineers are the requirements imposed by governing bodies. In meeting with the objectives of the control system, it is imperative that the impact on the environment is non-existent, or at the very least, minimal. For this goal to be achieved an optimal control strategy needs to be implemented. The question from which has stemmed many research work has been, “What control strategy would best fit this purpose?” This is also the premise for this research work.

Optimization deals with how to do things in the best possible manner. For obvious reasons, this concept has received an increasing amount of attention in recent years (Wismer and Chattergy, 1978). Optimal control theory has as its objective the maximization of the return from, or the minimization of the cost of, the operation of physical, social, and economic processes. Simply put, it determines the control signals that will cause a process to satisfy the physical, legal, environmental, and economic constraints, and at the same time minimize (or maximize) some performance criteria (Kirk, 1970).
1.4.1 Introduction to Model Predictive Control (MPC)

A control strategy that could be said to be optimal is Model Predictive Control (MPC). This is a model based control strategy whereby a model of the plant to be controlled is used in the design of the control algorithm. It utilizes the available model of the system in its design to predict the future behaviour of the process. It is important to note that the model of choice should accurately capture all the inherent properties of the plant.

Model Predictive Control (MPC) was developed in the late seventies, and it generated a huge research interest. The developments till date have yielded far reaching results (Camacho et al., 2010). At the moment it is said to be the most widely accepted multivariable control algorithm in the industries (Rangaiah et al., 2002). While MPC may be suitable for almost any kind of problem, it displays its main strength when applied to problems with:

- A large number of manipulated and controlled variables.
- Constraints imposed on both the manipulated and controlled variable.
- Changing control objectives and/or equipment failure.
- Time delays.

Some of the more common implementations of MPC are Dynamic Matrix Control (DMC), Model Algorithmic Control (MAC), and Generalized Predictive Control (GPC). While these algorithms differ in certain details, the main ideas behind them are very similar. Ideas common to all MPC implementations basically include:

- The use of a model to predict the process output at future time instants.
• Calculation of a control sequence by minimizing an objective function
• Receding strategy so that at each instant the horizon is displaced towards the future.

This involves the application of the first control signal of the sequence calculated at each step.

In its unconstrained form, MPC is closely related to Linear Quadratic (LQ) optimal control. When constraints are considered, MPC becomes an optimization problem, which is solved online in real-time at each sampling interval. This will be reviewed in more details in subsequent chapters.

1.5 Statement of the Problem

The packed bed tubular reactor is widely used in the process industries. One of the challenges of this reactor, however, is maintaining the temperature within the reactor. Temperature management is crucial when designing a catalytic tubular reactor for chemical reactions because hot spots in packed-bed tubes affect conversion, selectivity, and lifespan of catalysts (Kim et al., 2012). However, hot spot development is a phenomenon prevalent among exothermic reactions. Endothermic reactions, on the other hand, require the application of heat to move the reaction in the forward direction. In order to avoid over-heating or under-heating, temperature management using appropriate control strategies is of the essence. This would help maintain optimum temperature within the reactor, thereby enhancing product yield.

The work of Akpan et al. (2007), as shown in Figure 1.3 below, simulated an ideal environment where they used a furnace with a single heating zone for catalyst
performance evaluation. This posed an environment where the parameters of temperature could be controlled. In the real world however, this is not the case. Hot spots could develop; there could also be pressure drop along the catalyst bed that would inevitably affect the physical and chemical properties of the catalyst as well as the conversion.

Figure 1.3 Schematic diagram of the experimental setup for the $CO_2$ dry reforming of methane using packed bed tubular reactor (Akpan et al., 2007)

1.6 Scope and Objective of Research Work

The purpose of this research work is to design and implement a nonlinear model predictive control strategy for the carbon dioxide reforming of methane. This strategy will replicate a “real world” environment as would be experienced in the process.
industries. A fuzzy logic based model of the plant will be utilized in the design of the control strategy. This has the advantage of giving a linear approximation of the nonlinear plant. Specifically, an Adaptive Neuro-Fuzzy Inference System (ANFIS) model of the plant is used in the design of the controller. This modeling framework combines the network-based optimization scheme of neural networks together with the ability of fuzzy logic to model uncertainties to derive the linear approximation of the nonlinear model.

1.7 Organization of Thesis

This thesis is presented in five chapters. Chapter 2 delves into a literature review of Model Predictive Control. It highlights the various industrial applications of this control strategy, especially in the process industries. Also reviewed in the same chapter are the various models applied in the implementation of this control strategy. Chapter 3 explains the principles and theories applied in this research work (methodology). The developed control strategy applied to the chemical reactor is simulated under various conditions and the analysis, results, and relevant discussions are presented in Chapter 4. The final chapter presents the conclusions drawn from this research work as well as recommendations for future studies.
2. LITERATURE REVIEW

2.1 Hydrogen Production

Hydrogen, as an energy vector, is projected to be the optimum solution to our energy crisis (Kothari et al., 2008). It is the simplest element on earth with an atom consisting of only one proton and one electron. Hydrogen is the most plentiful element in the universe. Despite its simplicity and abundance, hydrogen does not occur naturally as a gas on the Earth. It is always combined with other elements. Water, for example, is a combination of hydrogen and oxygen. Hydrogen is also found in many organic compounds, notably the “hydrocarbons” that make up many of our fuels such as gasoline, natural gas, methanol and propane (Momirlan and Veziroglu, 2005).

As a result of the occurrence of hydrogen in combination with other elements, it has to be extracted, in order for it to be used for other purposes, like a fuel. Hydrogen can be extracted in a number of ways, such as electro-chemical processes, thermo-chemical processes, photo-chemical processes, photo-catalytic processes, or photo-electrochemical processes (Momirlan and Veziroglu, 1999, 2002). The major methods of hydrogen production from various energy sources are given in Figure 2.1 below.
Hydrogen is a secondary form of energy, produced by using three different energy-supply system classes, namely, fossil fuels (coal, petroleum, natural gas, and as yet largely unused supplies such as shale oil, oil from tar sands, natural gas from geopressed locations, etc.), nuclear reactors including fission reactors and breeders, and finally renewable energy sources (including hydroelectric power, wind power systems, ocean thermal energy conversion systems including biomass production, photovoltaic energy conversion, solar thermal systems, etc.) (Kothari et al., 2008).

Currently hydrogen is mostly produced by steam reforming of methane (Kothari et al., 2006). Renewable and nuclear systems can also produce hydrogen from water.
using thermal or electrolytic processes. Bio-hydrogen production by anaerobic fermentation from renewable organic waste sources has been found to be a promising method for the recovery of bio-energy (Han and Shin, 2004). In this method, anaerobic bacteria use organic substances as the sole source of electrons and energy, converting them into hydrogen (Balat, 2009).

The global hydrogen production presently relies heavily on processes that extract hydrogen from fossil fuel feedstock as shown in Figure 2.2 below. One of the drawbacks of hydrogen production from fossil fuel feedstock is a phenomenon known as carbon footprint (carbon dioxide emission). Transition from fossil fuels to renewable energy resources would have to be gradual. In the meantime while fossil fuels are still being burnt, the research interest would be the optimal production of hydrogen with minimal or no carbon dioxide emission to the environment.

Figure 2.2 Feedstock used in the current global production of hydrogen (Kothari et al., 2006)
It can be seen from Figure 2.2 that 96% of the hydrogen produced globally is produced directly from fossil fuels, and about 4% is produced indirectly by using electricity generated through fossil fuels (Boessel et al., 2003).

2.2 Hydrogen Production from Conventional Sources

In 2005, 48% of the global demand for hydrogen was produced from steam reforming of natural gas, 30% from oil/naphta reforming from refinery/chemical industrial off-gases, 18% from coal gasification, 3.9% from water electrolysis and 0.1% from other sources (Ewan and Allen, 2005, Kothari et al., 2006.). With the prospects of hydrogen being an important carrier and storage medium of energy, efforts should be made to reduce the amount the carbon dioxide emitted into the atmosphere from conventional sources (Lemus and Duart, 2010).

2.2.1 Natural Gas Steam Reforming

As previously stated, much of the hydrogen produced globally is obtained from natural gas, which is mostly made up of methane. Industrially, hydrogen is produced through the catalyzed steam reformation of methane (Momirlan and Veziroglu, 1999). Steam reforming of methane is currently the least expensive method of producing hydrogen, and is used for about half of the world’s production of hydrogen (Levent et al., 2003, Balat, 2009). It is an endothermic reaction, thereby requiring the addition of heat to promote the forward reaction. The temperature range for the reaction varies depending on the industrial or experimental setup. In all cases reviewed in the literature, the reactions required temperatures not less than 773 K. Levent et al., (2002) carried out the reforming process in a temperature range of 873 K – 1113 K
while Balat, (2009) reported reaction temperature in the range of 773 K – 1223 K. In all cases however, the reaction was catalyzed by a Nickel catalyst.

The steam reforming of methane is basically a two-step process with hydrogen as the product. At elevated temperature and pressure, methane is reformed catalytically to produce synthesis gas (syngas), which is a mixture of hydrogen and carbon monoxide. This reaction is then followed by a catalytic shift reaction to combine the carbon monoxide with water to produce more hydrogen. This catalytic shift reaction is known as the water gas shift (WGS) reaction.

Reforming reaction: $CH_4 + H_2O \leftrightarrow CO + 3H_2 \Delta H_{298}^{\circ} = +206kJ/mol$ (2.1)

Water Gas Shift reaction: $CO + H_2O \leftrightarrow CO_2 + H_2 \Delta H_{298}^{\circ} = -42kJ/mol$ (2.2)

Purification of the hydrogen produced is carried out in a pressure swing absorption (PSA) unit (Pilavachi et al., 2009). Balat, (2009) reported the purity of hydrogen to be as high as 99.99%. A major disadvantage of the steam reforming of methane is that hydrogen production is accompanied by the emission of large quantities of CO$_2$ because fossil fuels are used as the raw materials and also for providing heating (Kothari et al., 2006).

2.2.2 Partial Oxidation of Hydrocarbons

Catalytic partial oxidation of (POX) of hydrocarbons is another of the many methods for producing hydrogen for mobile and stationary applications (Newson and Truong, 2003). In this process, various hydrocarbons (e.g. ethane $C_2H_6$) are oxidized in order
to produce synthesis gas (Pilavachi et al., 2009). After the synthesis gas production, a WGS reaction takes place to reduce the carbon monoxide as shown in equation 2.3 below.

\[ \text{C}_2\text{H}_6 + \text{O}_2 \leftrightarrow 2\text{CO} + 3\text{H}_2 \]  

(2.3)

All kinds of gaseous and liquid fuels can be used in these reactions. Therefore, it is mostly used for processing high-boiling and high-sulphur-containing raw materials such as heavy oil, or petroleum refinery residual oil (Kothari et al., 2006). Purification of the hydrogen is carried out in a PSA unit. Just like in the steam reforming of methane, a drawback of this system is the emission of \( \text{CO}_2 \).

### 2.2.3 Coal Gasification

The coal gasification process is used in commercial production of synthesis gas, as a means towards the clean use of coal (Gnanapragasam et al., 2010). The reaction mechanism of coal gasification is similar to that of the partial oxidation of heavy oils. The basic reaction for the gasification of coal is as shown below:

\[ \text{CH}_{0.8} + 0.6\text{O}_2 + 0.7\text{H}_2\text{O} \leftrightarrow \text{CO}_2 + \text{H}_2 \]  

(2.4)

Hydrogen production by coal gasification can occur in either of 2 ways: the synthane process and the \( \text{CO}_2 \) acceptor process. For the first process, coal reacts with steam at high temperature and pressure. Gaseous products of carbon monoxide, carbon dioxide and hydrogen are produced with a small amount of methane (Kothari et al., 2006,
Balat, 2009). After purification, the final gas is 97 – 98% pure. The acceptor process involves lime, which is introduced with coal when it reacts with steam. The carbon dioxide produced is removed by the lime as calcium carbonate. Once the carbon dioxide has been removed, the shift reaction can occur in the main reactor thereby eliminating the need for an external shift reactor. A major downside of this process is that it produces more carbon dioxide than the other technologies.

2.3 Hydrogen Production from Biomass and Renewable Sources

Hydrogen produced from renewable sources would most likely play an important role as an energy carrier in the future energy supply. Due to the finite life of fossil fuels and the global environmental damage caused by fossil fuels, the world has to switch gradually to renewable energy sources such as water, wind, and sun. Electrolysis is often considered as it is the only process that does not rely on fossil fuels. Another advantage of using electrolysis is the high product purity it gives. Moreover it is feasible on both small and large scales. Electrolysis only has a carbon footprint when fossil fuels are used to generate the electricity needed for the process. Production of hydrogen from sustainable harvested biomass, solar energy, or wind energy will considerably reduce the production of emissions. However, carbon dioxide emissions can be reduced to zero, if the process of distributed electrolysis is applied. Distributed electrolysis involves using electricity from wind and solar energy.

**Solar power:** Solar energy is a clean, renewable energy source and has attracted much attention as an alternative to fossil energy for future use.
**Wind power:** Hydrogen can be produced by electrolysis using wind turbines as the source of electricity. Of all the renewable sources, using wind turbine-generated electricity to electrolyse water has the greatest potential for producing pollution-free hydrogen.

**Hydropower:** electricity production from hydropower is also the source of hydrogen production. This electricity is further used in the splitting of water molecules to produce emission-free hydrogen.

From discussions above it can be clearly seen that hydrogen production from hydrocarbons is well established and most commonly used. This poses the question of how hydrogen production process can be made cleaner. In an attempt to address the issue of pollution, a method of hydrogen production has emerged. This process is the Carbon Dioxide Reforming of Methane (CDRM).

### 2.4 Carbon Dioxide Reforming of Methane

As the major constituent of natural gas, methane has attracted increased attention in recent years. It is considered an important raw material for the production of higher hydrocarbons and petrochemicals due to the predicted oil shortage in the future. However, since methane is not very reactive, its direct conversion to useful hydrocarbon products is well known to be a difficult process. At present, most commercial processes for conversion of methane to useful products are indirect processes, where methane is first converted to synthesis gas and subsequently converted to a variety of products by the Fisher-Tropsch process. The principal routes
for the conversion of methane to synthesis gas include steam reforming, partial oxidation, and carbon dioxide reforming. Each of these processes has advantages and disadvantages (Li et al., 2010, Li et al., 2008).

Among the three methods, the reforming of methane by carbon dioxide to produce synthesis gas has drawn special attention due to environmental considerations. The reaction proceeds according to the following equation:

\[ \text{CH}_4 + \text{CO}_2 \leftrightarrow 2\text{CO} + 2\text{H}_2 \Delta H_{298} = 247 \text{ kJ/mol} \]  

(2.5)

By this reaction, two of the most abundant carbon-containing greenhouse gases, carbon dioxide and methane may be converted to synthesis gas, which may then be used for the production of petrochemicals and synthetic fuel alternatives (Akpan et al., 2007, Arbag et al., 2010, Barroso-Quiroga and Catro-Luna, 2009). The major challenge to hydrogen production by CDRM is usually catalyst deactivation due to carbon deposition on the catalyst surface (Arbag et al., 2010, Fidalgo et al., 2008, Barroso-Quiroga and Catro-Luna, 2009). The primary focus in the research community has been about improving the catalysts used in the CDRM process (Akpan et al., 2007, Fidalgo et al., 2008).

2.5 Tubular Reactors and Temperature Control

The CDRM process, an endothermic process, requires an external heat source to provide the thermal energy for the reaction. This reaction is carried out in a reformer or a plug flow reactor (PFR). Tubular reactors are of great importance in the chemical
industry. This is mainly due to their efficient product extraction, large flow velocities, and high yields (Aguilar et al., 2002). These reactors, over the years, have been an active area for both academic and industrial research (Wu and Huang, 2003). They present an interesting challenge for process control engineers because the reactors are modeled as distributed parameter systems. The composition and temperature profiles of the system vary with time and space. Moreover, as with many other chemical processes, they exhibit strong nonlinearities and non-stationary characteristics. This, in addition to the difficulty in measuring the controlled variable, for example conversion, has made this a peculiar control problem (Chen and Sun, 1991).

Several attempts, over the years, have been made at resolving control problems with the most problematic being temperature control (Luyben, 1999). Controlling the temperature of the reactor is a very critical determinant of the reactor performance and catalyst life (Vernon et al., 2009). The presence of excessive temperatures within the reactor has detrimental consequences on the operation of the reactor. These excessive temperatures could lead to catalyst deactivation, undesired side reactions, and thermal decomposition of the products (Karafyllis and Daoutidis, 2002). Consequently, control strategies that regulate the magnitude of the ambient temperature are of paramount importance. To achieve this, closed-loop control strategies have been used to modulate the heat input rate based on a comparison between a set point temperature and a temperature measurement. Karafyllis and Daoutidis (2002) were able to suppress the magnitude of hot spots in a plug flow reactor by manipulating the jacket coolant temperature. They achieved this by
deriving a nonlinear control law, which guaranteed that the temperature of the hot spot remained below a pre-specified bound. The designed controller was shown to exhibit excellent robustness with respect to modeling and measurement errors. Chen and Sun (1991), applied adaptive inferential control strategy to control a packed bed reactor. Control was accomplished by using secondary measurements of temperature along the reactor length. In this case, the measured variable was not accessible. In order to achieve control, estimators based on the nonlinear models of the plants were used to accurately predict the composition (Chen and Sun, 1991). To further emphasize the importance of temperature control in the chemical processes, some studies have been focused on determining the optimal temperature profile for maximizing product yield (Nouralishahi et al., 2008, Logist et al., 2007, Logist et al., 2008).

2.6 Applications of Model Predictive Control (MPC) Strategy
Model predictive control (MPC) is a control strategy that has received wide spread acceptance in the process industry. It is a model based control strategy whose underlying rationale is to transform the control problem into an optimization problem. At every sampling instance, a sequence of future control values is computed by solving a finite horizon optimal control problem (Scattolini, 2009).

The current interest of the process industry in MPC could be traced back to the late 1970s. A set of papers by Richalet et al., (1978) and engineers from Shell in 1979 described successful applications of MPC. In 1978 Richalet et al. wrote about successful applications of “Model Predictive Heuristic Control”, and in 1979
engineers from Shell outlined “Dynamic Matrix Control” and reported applications to a fluid catalytic cracker (Garcia et al., 1989).

The importance of MPC is that it provides a general control scheme where material and/or energy conversion equipment and the control devices can be considered as a whole in the design of the control systems (Tsai et al., 2002). The basic idea of MPC is to use a model to predict the future output trajectory of a process and compute a series of controller actions to minimize the difference between the predicted trajectory and a user-specified one, subject to constraints (Tsai et al., 2002).

2.6.1 Application of MPC in Robotics

The application domain for MPC is very diverse because of its capacity to incorporate various practical control objectives and requirements. MPC, though a control strategy that is computationally intensive, has found application in both processes with slow dynamics and in processes with fast dynamics (Richalet, 1993). Temurtas et al. (2006) successfully applied MPC to the control of robotic manipulators. Robotic manipulators are examples of systems with fast dynamics, and are mostly employed in the manufacturing industries. They have dynamic characteristics that are highly nonlinear functions of position and velocity. To overcome the challenge posed by the nonlinearities in the system, the researchers used a neural network based model of the robotic manipulator in the control design. It was reported that the motion of the manipulator was very smooth (Termutas et al., 2005). In another study, MPC was applied in the control of car-like mobile robots. These robots are mostly used in ports, planet exploration, nuclear waste cleanup, agriculture, and mining. For this study
MPC was employed in path tracking for the robots. Similarly as in the first case, a neural network was used to model the complex nonlinear dynamics of the system (Gu and Hu, 2002).

2.6.2 Application of MPC in Medicine

In a study carried out in the medical field, an MPC algorithm was developed for a bio-inspired flexible probe. These needles are used for minimally invasive surgery (MIS) for the diagnosis and treatment of a variety of medical pathologies. For this study a tracking error model was modified so that the nonlinear kinematic model of the probe was linearized, thereby enhancing fast computations (Ko and Baena, 2012). In another study, adaptive MPC was used in the maintenance therapy for childhood acute lymphoblastic leukemia. The adaptive MPC was used for personalized patient care by using routine measurements of red blood cell mean corpuscular volume as a substitute for the active drug metabolite concentration. According to Noble et al., (2010) a clinically relevant mathematical model was used to describe the patient response to the chemotherapeutic drug 6 – mercaptopurine, with some model parameters being patient specific. This study concluded that the use of the applied model with the adaptive MPC could be a valuable tool for creating a personalized treatment strategy that was both safe and effective (Noble et al., 2010). Mahfouf et al. (2003) applied the MPC algorithm in administering anesthetic drug during surgery. As an objective of this study it was required that the right depth of anesthesia be maintained during surgery. After clinical evaluation trials, the anesthetist concluded that good anesthesia was achieved and the patients had good recovery after the
operation (Mahfouf et al., 2003). In another medically related study, MPC was applied in the control of glucose concentration for Type 1 diabetic patients. The algorithm was able to establish control by using subcutaneous insulin delivery and subcutaneous continuous glucose monitoring. This study was able to highlight the crucial role of the anticipative feedforward action driven by the meal announcement information. The control strategy based on a linear model was able to achieve good results though it was suggested that better results could be achieved, if a nonlinear model was applied (Magni et al., 2009). The MPC algorithm was applied yet again in another study by Mohammed et al. (2012), for the functional electrical stimulation of knee joint quadriceps muscles. This study is beneficial to patients with spinal cord injuries who may have lost total or partial control of the lower limb. In this study, appropriate stimulation patterns were computed as a function of the desired lower-limb knee joint movements. The proposed controller showed satisfactory results in terms of regulation, stability, and robustness with respect to external disturbances.

2.6.3 Application of MPC in Process Systems

As stated earlier, the origins of the MPC algorithm can be traced back to the chemical process industry, as such the industry has been a major benefactor of this algorithm. This industry is dominated by processes that have slow dynamics, and MPC lends itself to these processes. In the chemical process industry, the drive to reduce operating costs and develop new markets has frequently emphasized improvements in product quality, better use of energy resources and reduced environmental emissions (Temeng et al., 1995). According to Temeng et al., (1995) these objectives, in turn,
have placed stringent requirements on the available process control systems. They further reported that these control systems must usually cope with multi-variable process interactions, constraints on manipulated and controlled variables, as well as time delays and other problematic dynamic characteristics. Model predictive control algorithms have been recognized as effective tools for handling the difficult control problems in this industry (Temeng et al., 1995).

MPC schemes derive some of their industrial appeal from their ability to handle input and output constraints, time delays, non-minimum phase behavior, and multi-variable systems (Temeng et al., 1995). It is a generally accepted fact that the most effective way to generate the most profit out of the plants while responding to marketplace variations with minimal capital investment is by the integration of all aspects of automation of the decision making process (Garcia et al., 1989). The petro-chemical industry is one of the industries that has benefitted from the implementation of MPC in its control structure, and it is characterized by very dynamic and unpredictable marketplace conditions. In a study carried out by Yuzgec et al. (2010), the MPC algorithm was applied in refinery scheduling of crude oil unloading, storage, and processing. Scheduling is the process of allocating scarce resources in an optimal manner, to maximize production and minimize waste. The goal of scheduling is to determine the most suitable time to perform each operation in accordance with the relationships between the production process and the capacity limitations of shared resources (Yuzgec et al., 2010).
2.7 Models in the MPC Strategy

A major challenge with MPC implementation in the process industry has always been with selecting a suitable model for the process. A suitable model is one, which is capable of accurately capturing the process dynamics so as to precisely predict the future outputs as well as being simple to implement and understand (Camacho and Bordons, 1999). The process industry, as well as most engineering fields, is characterized by systems that are highly nonlinear with parameters that vary in time and space. This is evidenced in the decades of research leading to a great deal of literature on distributed parameter systems scattered in a wide spectrum (Padhi and Ali, 2009). Solving for these distributed parameter systems is a time consuming procedure and this contradicts the aim of designing controllers. Process controllers should be able to respond to disturbances in the process system in a timely manner. Unfortunately MPC is a time consuming optimization procedure and using a partial differential equation (PDE) model of the process increases the time it takes for the optimization to be done. Despite this fact, there are a large number of real applications described by PDE models. Moreover, as modeling becomes more and more accurate the use of complex nonlinear PDE models is increasing. However, only few studies have been devoted to the control of processes explicitly characterized by PDE models, especially the nonlinear case. This has been due to the complex nature of the control problem. On the bright side various methods have been proposed for control of such distributed parameter systems, but no general framework is in place yet (Dufour et al., 2003, Dufour et al., 2004).
2.7.1 MPC Strategy Based on Linearized Models of the Process

Researchers designing control systems for processes that are nonlinear and with parameters varying spatiotemporally have considered alternate models. In some cases, to deal with the nonlinearities present in the process, the process is linearized around the operating point then the linear model is applied in the control design. In a study by Ozkan et al. (2003), multiple piecewise linear models of a solution copolymerization reactor was used in the MPC strategy. It was shown that this approach had the ability to rapidly transition the process between different operating points. However, MPC algorithms, which use a linear process model in prediction and optimization, would deteriorate when they are applied to processes with strong nonlinearities (Seki et al., 2001).

2.7.2 Nonlinear MPC Strategies

Nonlinear model predictive control possesses a strong potentiality in improving control, and in the operation of nonlinear processes. The underlying principle of nonlinear model predictive control (NMPC) is the same as linear model predictive control (LMPC) with the exception that the model describing the process dynamics is nonlinear. However, from implementation viewpoints, it poses some technical problems, which are associated with computational burdens. The application of NMPC, in which a nonlinear programming problem has to be solved online, requires formidable efforts in order to calculate control actions within fixed sampling time (Seki et al., 2001).
2.7.3 Neural Network Based MPC

Studies have shown that the integration of nonlinear models in MPC designs for the process industry has been very successful. Neural networks have been used extensively for a number of chemical engineering applications such as sensor data analysis, fault detection, and nonlinear process identification because of its good modeling capabilities. Models based on neural networks have been extensively applied to the predictive control strategy (Hussain, 1999). In a study by Hosen et al. (2011), a neural network model of a polystyrene batch reactor was used in developing an MPC strategy for the batch reactor. This control strategy was observed to be better than the conventional PID controller. The NN-MPC resulted in smoother controller moves and less variability (Hosen et al., 2011). In another study, a neural network model of a yeast fermentation process was used in the design of an MPC strategy (Lawrynczuk, 2011). The main problem with using neural network as a model is that its performance deteriorates considerably when applied to data outside the range it was trained with. It has a poor extrapolation property (Arefi et al., 2008). To avert this, neural networks are usually applied in conjunction with other modeling and identification techniques. In a study by Arefi et al. (2008), a neural network based wiener model of a plug flow reactor was used for identification and predictive control. Simulation results from the identification phase proved that the identified model was valid.
2.7.4 Soft Computing and Control

Soft computing techniques play an important part in the modeling of these processes. Soft computing research is concerned with the integration of artificial intelligence tools (neural networks, fuzzy technology, evolutionary algorithms, etc.) in a complementary hybrid framework for solving real world problems (Denai et al., 2007). In a study by Golob and Tovornik (2008), a neuro-fuzzy ARX model was developed for a nonlinear process based on an input-output data set. From their work they concluded that the method was accurate to a high degree and had linguistic rules that were interpretable. In another study, a recurrent fuzzy neural network was applied to industrial processes. This control strategy was illustrated using a laboratory-scale liquid-level process and in a variable-frequency oil-cooling process. It was concluded that this control strategy was useful and pragmatic for achieving set-point tracking, either in the presence or absence external disturbances and load changes (Lu and Tsai, 2007).

2.7.4.1 Adaptive Neuro-Fuzzy Inference System (ANFIS) Modeling and Identification

Another modeling and identification technique commonly applied is Adaptive Neuro-Fuzzy Inference System (ANFIS). This is a neuro-fuzzy modeling technique that applies a hybrid learning algorithm combining back propagation (BP) and least square estimate (LSE) during the modeling process. This hybrid learning algorithm is adopted to identify the linear and nonlinear parameters in the ANFIS model. An advantage of neuro-fuzzy modeling is that it presents an easy way to derive successful
models. ANFIS is a modeling algorithm with very diverse applications. Esfahanipour and Aghamiri (2010); Chang et al. (2011) successfully applied ANFIS in performing stock market analysis. The ANFIS model was used in stock price prediction, and it was able to accurately forecast the stock prices (Esfahanipour and Aghamiri, 2010; Chang et al., 2011). Boyacioglu and Avci (2010) also concluded that ANFIS provides an alternative model for stock market predictions, and can be a useful tool for economist and practitioners dealing with the forecasting of the stock price index return based on their Istanbul Stock Exchange study. In another study conducted by Chen (2011), an ANFIS model was used for predicting business failure prediction using particle swarm optimization and subtractive clustering.

In the field of telecommunications, Alotaibi et al. (2008) developed a robust prediction model using ANFIS based on Terrestrial Trunked Radio (TETRA) outdoor Radio Frequency (RF) measurements. This model was used to predict the strength of the wireless signal received by wireless devices. In this study carried out in Saudi Arabia, it was observed that the ANFIS prediction model outperformed the predictors based on empirical models, and it was marginally better than Radial Basis Function – Neural Network (RBF-NN) predictors (Alotaibi et al., 2008).

In the process industry, ANFIS has been applied in a diverse number of applications. In a study conducted by Han and Zhao (2011), a dynamic control model was designed for a basic oxygen furnace (BOF) steel making process. This model was based on ANFIS and a robust relevance vector machine. An ANFIS classifier was used to determine whether the coolant should be added or not then an ANFIS regression
model was used to calculate the amount of oxygen and coolant required. Simulation on industrial data showed that the dynamic control model yielded good results (Han and Zhao, 2011). In another study ANFIS was used, in addition to a Finite Element Model (FEM), to predict the wafer surface non-uniformity in a chemical mechanical polishing (CMP) process. The data ANFIS was trained from was obtained under several conditions of the carrier load, the pads elastic modulus, and thickness by using the developed finite element model for CMP. Lau et al. (2010) applied ANFIS for the fault diagnosis of the polypropylene production process (UNIPOL PP). The emphasis of this study was on fast and accurate diagnosis, multiple fault identification and adaptability. Simulation results showed that the method effectively diagnosed different fault types and severities. It was also observed that it had a better performance compared to a conventional multivariable statistical approach based on principal component analysis (PCA). In addition, it was shown that the method was simple to apply, robust to measurement noise, and it was able to rapidly discriminate between multiple faults occurring simultaneously (Lau et al., 2010).

Mullai et al. (2011) applied ANFIS for modeling an anaerobic hybrid reactor (AHR). The hybrid reactor was used in treating penicillin – G waste water at the ambient temperatures of 30 – 35 °C for 245 days in three phases. It was concluded that the ANFIS model was well performed in predicting the performance of the AHR (Mullai et al., 2011). In a study conducted by Wu et al. (2008), an ANFIS model was developed to study different flows effect on the performance of solid oxide fuel cell (SOFC). The validity and accuracy of this model was tested by simulations, and the
results of these simulations revealed that the obtained ANFIS model efficiently approximated the dynamic behaviour of the SOFC stack (Wu et al., 2008).

2.8 MPC Strategy Based on ANFIS Models

In a study conducted by Dovzan and Skrjanc (2010), predictive functional control strategy was designed for a semi-batch reactor. An ANFIS model was used in connection with a fuzzy predictive functional control to construct an adaptive fuzzy predictive functional controller. This control strategy was tested on a nonlinear, time-varying, semi-batch reactor process. The simulation indicated that using this approach could improve the control of the semi-batch reactors. In another study carried out by Escano et al. (2009), an MPC control strategy, based on an ANFIS model, was applied to a thermal batch process. The researchers reported that the low computational cost associated with this strategy made them suitable for implementation in industrial Programmable Logic Controllers (PLC) (Escano et al., 2009). This work shares some similarities with the research work being presented in this thesis. In both studies emphasis is placed on the temperature control of the processes. However, instead of a batch reactor, this work was focused on a plug flow reactor. The plug flow reactor is a distributed parameter system with nonlinear dynamics thus, the need for a neuro-fuzzy modeling scheme. The neuro-fuzzy model will then be used in an MPC scheme for designing a control strategy for the process. The procedure is illustrated in the flow chart below:
Figure 2.3 Procedure for the design of an MPC strategy

This procedure will be discussed in greater details in the following chapters.
3. METHODOLOGY

3.1 Model Development

As stated in Section 2.8 of Chapter Two, this research work is very similar to studies conducted by Dovzan and Skrjanc (2010) and Escano et al., (2009). Both groups of researchers used batch reactors in their respective studies, and much emphasis was placed on the temperature control of the processes. However, instead of a batch reactor, a plug flow reactor was employed in this research work. This reactor guarantees efficient product extraction, large flow velocities, and high yields (Aguilar et al., 2002). In addition, it presents different sets of challenges other than that of the batch reactor. Plug flow reactors are highly nonlinear, with parameters that vary spatiotemporally.

Akpan et al. (2007) in their research work modeled the reactor based on the set of differential equations as presented below. Equation 3.1 is the mass balance equation and equation 3.2 is the energy balance equation.

\[ u_z \frac{\partial C_i}{\partial z} = D_{eff} \frac{\partial^2 C_i}{\partial r^2} + \frac{D_{eff}}{r} \frac{\partial C_i}{\partial r} + D_{eff} \frac{\partial^2 C_i}{\partial z^2} + \rho_B v_i r_j \]  

(3.1)

\[ \rho_g C_p u_z \frac{\partial \tau(r,z)}{\partial z} = \lambda_{eff} \frac{\partial^2 \tau(r,z)}{\partial r^2} + \lambda_{eff} \frac{\partial \tau(r,z)}{\partial r} + \lambda_{eff} \frac{\partial^2 \tau(r,z)}{\partial z^2} + (\Delta H_j) \rho_B v_i r_j (r, z) \]  

(3.2)

where \( u_z \) = superficial velocity (km/h)

\[ C_i = \text{feed concentration of species (mol/m}^3) \]

\[ D_{eff} = \text{effective diffusivity (m}^2/\text{h)} \]

\[ r = \text{radius of reactor (m)} \]
\[ \rho_B = \text{bulk density (kg/m}^3) \]

\[ \nu_i = \text{stoichiometric coefficient of species } i \]

\[ r_j = \text{rate of reaction} \]

\[ \rho_g = \text{gas density (kg/m}^3) \]

\[ C_p = \text{heat capacity (kJ/kg.K)} \]

\[ T(r, z) = \text{reactor temperature (K)} \]

\[ \lambda_{eff} = \text{effective thermal conductivity (kJ/m.s.K)} \]

\[ \Delta H_j = \text{heat of reaction (kJ/kmol)} \]

This set of equations were expressed for each component species \( i \) in the radial and axial directions in the cylindrical coordinate system by considering the mass and energy balance of the reactor. For this study, the reactor was run at steady state, as expressed by the mathematical model. In order to design a control strategy for this process, the dynamic behavior of the process would need to be studied. This would require modeling the time dependency of the dependent parameters.

As stated earlier this research work is based primarily off the initial work by Akpan et al. (2007) who conducted kinetic, experimental modeling, and simulation studies of CDRM in a packed bed tubular reactor. However, my focus is on the design of an appropriate control strategy to maintain the desired conditions within the reactor to ensure optimal methane conversion. In my case, a plug flow reactor was utilized. The plug flow reactor is a distributed parameter system with nonlinear dynamics thus, the need for simplification by a neuro-fuzzy modeling scheme. This derived neuro-fuzzy
model was then used in an MPC algorithm for designing a control strategy for the process.

3.2 Chemical Reactor Modeling

In order to study a system or process, we need to set up a model of the system. The form and content of dynamic process models are on the one hand determined by the application of the model, and on the other by the available knowledge. The application of the model determines the external structure of the model while the available knowledge determines the internal structure. Dynamic models of processes can be used for simulation studies to get information about the process behavior. These models can also be used for control and optimization studies of the process. The process knowledge can be available as physical relationships or in the form of process data.

A model could be said to be an image of the reality, be it a process or a system. These models are developed based on a focused application.

Some of the areas where models are applied include

(1) Research and development
(2) Process design
(3) Planning and scheduling
(4) Process optimization
(5) Prediction and control
A model of the system is:

- A representation of the essential aspects of the system
- In a suitable (mathematical) form
- That can be experimentally verified
- In order to clarify questions about the system

As stated earlier, these models are usually based on physical fundamentals, conservation balances, and additional equations. To describe a process system, we need a set of variables that characterize the system and a set of relationships that describe how these variables interact and change with time. State variables are used to characterize a state and they include: density, concentration, temperature, pressure, and flow rate. These state variables can be derived from the conservation balances for mass, component, energy, and momentum.

This conservation is based on the fundamental physical law that mass, energy, and momentum can neither be created nor destroyed. This law applies to every defined system be it an opened system or a closed system. In process engineering, a system is usually a defined volume, process unit, or a plant (Roffel and Betlem, 2006).
As stated by Roffel and Betlem (2006), the mass in a closed system is always constant, and it is expressed mathematically as given below:

\[
\frac{\text{time rate of change of mass within system}}{\text{time}} = \frac{\text{net mass flow through boundary}}{\text{time}}
\]  

(3.3)

The component balance can be derived from the mass balance equation given above.

\[
\frac{\text{rate of change of component mass within system}}{\text{time}} = \frac{\text{net flow of component mass through boundary}}{\text{time}} + \frac{\text{net production of component mass through reaction within system}}{\text{time}}
\]  

(3.4)
The sum of the component balances of all components should agree with the mass balance. However, there can be a net production or consumption of a component within a system.

The energy balance is the result of the first law of thermodynamics. For an open system all kinds of environmental influences should be taken into account.

\[
\begin{align*}
\text{rate of change of internal + kinetic + potential energy within} &= \text{net flow of internal + kinetic + potential energy through boundary} \\
&+ \text{heat flow by conduction + radiation through boundary} \\
&+ \text{(electrical)heat from sources within system} \\
&+ \text{work done by system on environment}
\end{align*}
\]

(3.5)

The momentum balance is derived from Newton’s second law of motion, which states that a body will accelerate proportional to the force acting on it and in the direction of the force. It is given below:

\[
\begin{align*}
\text{net change of momentum in time} &= \text{net force working on boundary}
\end{align*}
\]

(3.6)

The momentum balance is the general case of this law allowing mass variation with time. It can be thought of as a dynamic force balance.

The state equations describe the behavior of the process with time. The accumulation is described by the terms on the left-hand side of the differential equation. The right-hand side contains the production and transport terms. The transport terms are caused
by differences compared to an equilibrium situation in pressure (momentum balance), temperature (energy balance) and composition (component balance).

3.2.1 The Plug Flow Reactor

The plug flow reactor is one of many idealized reactor models. It is a simplified picture of the motion of fluid, whereby all the fluid elements move with a uniform velocity along parallel streamlines. This perfectly ordered flow is the only transport mechanism accounted for in the plug flow reactor. In addition, the conditions at any point in the reactor are independent of time. The composition of the reaction mixture depends on the distance $L$ from the inlet point. In the plug flow reactor, it is assumed that no mixing occurs within the fluid, and the velocity profile is flat. Another assumption made is that both temperature and composition are uniform at any cross-section in the reactor.

The key assumptions made in deriving the mathematical model of a plug flow reactor are:

1. The plug flow in a tube is an ideal-flow assumption, in which the fluid is well mixed in the radial and angular direction.
2. The fluid velocity is assumed to be a function of the axial position in the tube.
3. Plug flow is often used to approximate fluid flow in tubes at high Reynolds number. The turbulent flow mixes the fluid in the radial and angular directions.
(4) Also in turbulent flow, the velocity profile is expected to be reasonably flat in the radial direction except near the walls.

### 3.2.2 The Packed Bed Tubular Reactor

The discovery of solid catalysts and their application to chemical processes in the early years of this century led to the many breakthroughs seen in the chemical industries. As a result of this, the industry has diversified and grown in a spectacular way. Notable improvements have been seen in the development of new or the rejuvenation of established process, mostly based on the use of solid catalyst (Froment and Bischoff, 1979).

The majority of the catalytic processes are carried out in a packed bed tubular reactor (Fixed Bed Reactor). The packed bed tubular reactor is a tubular reactor (Plug Flow Reactor) packed with solid catalyst particles. The catalysts in the reactor may be placed in one or more fixed beds (i.e., layers across the reactor) or may be distributed in a series of parallel long tubes. The latter type of fixed-bed reactor is widely used in industry, and offers several advantages over other forms of fixed beds.

An advantage of the fixed-bed reactor is that for most reactions, it gives the highest conversion per weight of catalyst of any catalytic reactor. Another advantage is it provides large volumes of processed reactants. The disadvantages of the packed bed reactor are listed below:

1. The catalysts are highly prone to deactivation.
(2) The catalysts often require regeneration after relatively short period of operation. This may incur additional cost.

(3) It is difficult to control the heat-transfer in the catalyst bed.

(4) Some part of the catalyst surface remains unused as a result of the reaction system and the rate-controlling step.

Modeling of packed bed reactors can be grouped in two broad categories: pseudo-homogeneous and heterogeneous. Pseudo-homogeneous models do not account explicitly for the presence of catalyst, in contrast with heterogeneous models, which lead to separate conservation equations for fluid and catalyst (Froment and Bischoff, 1979). This classification was an attempt at justifying the degree of sophistication necessary in reactor modeling. The most widely used model in studies has been the pseudo-homogeneous one dimensional model. This model only considers transport by plug flow in the axial direction. Some type of mixing in the axial direction is superimposed on the plug flow so as to account for non-ideal flow conditions. Accounting for radial gradients in the model makes it a two dimensional model of the reactor. The basic model in the heterogeneous categories considers only transport by plug flow, but distinguishes between conditions in the fluid and on the solid.

3.2.2.1 The One Dimensional Reactor Model

The one dimensional model assumes that concentration and temperature gradients only occur in the axial direction. The only transport mechanism operating in this direction is the overall flow itself, and this is considered to be of the plug flow type. Assuming no axial mixing and negligible pressure drop along the length of the
reactor, the conservation equations may be written for the steady state and a single reaction carried out in a cylindrical tube.

**Material balance:**

\[-u_z \frac{dC_A}{dz} = \rho_B r_A\]  \hspace{1cm} (3.7)

**Energy balance:**

\[-u_z \rho_g c_p \frac{dT}{dz} = (-\Delta H) \rho_B r_A - 4 \frac{U}{d_t} (T - T_w)\]  \hspace{1cm} (3.8)

where the parameters are as defined: \(C_A\) is the concentration of the reactant; \(T\) is the average temperature within the reactor; \(T_w\) is the temperature of the cooling (heating) fluid; \(u_z\) is the superficial velocity of the fluid (e.g. gases) mixture; \(\rho_B\) is the bulk density of the fixed bed; \(\rho_g\) is the gas mixture density; \(r_A\) is the rate of reaction; \(c_p\) is the heat capacity of the gas mixture; \(\Delta H\) is the heat of reaction; \(U\) is the overall heat transfer coefficient; \(d_t\) is the diameter of the reactor.

### 3.2.2.2 The One Dimensional Reactor Model with Axial Mixing

The model discussed in equations 3.7 and 3.8 above, though sufficiently representative of the system and more convenient to use has been oversimplified. The flow in a packed bed reactor deviates from the ideal pattern because of variations in flow velocity and mixing effects due to the presence of packing. The assumption that the temperature is uniform in a cross section is an oversimplification. To address these concerns, the following models have been developed.
The mixing in the axial direction, which is due to turbulence and the presence of packing, is accounted for by superimposing an “effective” transport mechanism on the overall transport by plug flow. The flux due to this mechanism is described by a formula analogous to Fick’s Law for mass transfer or Fourier’s Law for heat transfer by conduction. The proportionality constants are “effective” diffusivities and conductivities (Froment and Bischoff, 1979).

**The steady state continuity equation for a component A may be written thus:**

\[
D_{ea} \frac{d^2 c_A}{dz^2} - u_z \frac{dc_A}{dz} - r_A \rho_B = 0
\]  

(3.9)

**The energy equation may be written as:**

\[
\lambda_{ea} \frac{d^2 T}{dz^2} - u_z \rho_g c_p \frac{dT}{dz} + ( - \Delta H ) r_A \rho_B - \frac{4\eta}{d} (T - T_w) = 0
\]  

(3.10)

The boundary conditions are:

For \( z = 0 \)

\[
u_z (C_{A0} - C_A) = -D_{ea} \frac{dC_A}{dz}
\]

\[
u_z \rho_g c_p (T_0 - T) = -\lambda_{ea} \frac{dT}{dz}
\]

For \( z = L \)

\[
\frac{dC_A}{dz} = \frac{dT}{dz} = 0
\]

where the parameters are as defined: \( T_0 \) is the feed temperature; \( z \) is the axial direction of the reactor; \( L \) is the length of the reactor; \( D_{ea} \) is the effective diffusivity in the axial direction; \( \lambda_{ea} \) is the effective axial conductivity.
3.2.2.3 The Two Dimensional Pseudo-Homogeneous Models

In the one-dimensional model, the resistance to heat and mass transfer in the radial direction was neglected, thereby uniform temperatures and conversions were predicted in a cross section. This simplification has serious implications when reactions with pronounced heat effect are involved. This prompts the need for a model that predicts the detailed temperature and conversion pattern in the reactor. Thus, the focus of the design would be avoiding eventual detrimental over-temperatures in the axis.

The following models apply the effective transport concept to formulate the flux of heat or mass in the radial direction. This flux is superimposed on the transport by overall convection, which is of the plug flow type (Froment and Bischoff, 1979).

The steady state continuity equation for a component A may be written thus:

\[
\frac{\partial}{\partial \tau} \left( D_{er} \frac{\partial C}{\partial \tau^2} + \frac{1}{r} \frac{\partial C}{\partial r} \right) + D_{ea} \frac{\partial^2 C}{\partial z^2} - u_z \frac{\partial C}{\partial z} - \rho_B r_A = 0
\]

(3.11)

The energy equation may be written as:

\[
\frac{\lambda_{er}}{\partial_\tau} \left( \frac{\partial^2 T}{\partial \tau^2} + \frac{1}{r} \frac{\partial T}{\partial \tau} \right) + \lambda_{ea} \frac{\partial^2 T}{\partial z^2} - u_z \rho g c_p \frac{\partial T}{\partial z} + \rho_B (-\Delta H) r_A = 0
\]

(3.12)

The boundary conditions are:

For \( z = 0 \)

\[ C = C_0, \quad T = T_0, \quad 0 \leq r \leq R_t \]
For $r = 0$ and $r = R_t$

$$\frac{\partial C}{\partial r} = 0$$

For $r = 0$ and all $z$

$$\frac{\partial T}{\partial r} = 0$$

For $r = R_t$

$$\frac{\partial T}{\partial r} = -\frac{\alpha_w}{\lambda_{er}} (T - T_w)$$

where the parameters are as defined: $C_0$ is the feed concentration; $r$ is the radial direction of the reactor; $D_{ea}$ and $D_{er}$ are the effective diffusivity in the axial and radial directions respectively; $\lambda_{ea}$ and $\lambda_{er}$ are the effective axial and radial thermal conductivity respectively and $\alpha_w$ is the heat transfer coefficient near the reactor wall.

3.3 The Simulation Model

For this work, the model used was comprehensive but left as simple as possible. This was achieved by modifying the model used by Akpan et al. (2007) to account for the time dependent response of the reactor. A one dimensional model of the process, taking note of the effective diffusivity and conductivity in the axial direction, was used in the system design. The proposed model is given below:

*The continuity equation for component A (methane, $CH_4$) may be written thus:*

$$\frac{\partial c_A}{\partial t} = -u_z \frac{\partial c_A}{\partial z} + D_{ef} \frac{\partial^2 c_A}{\partial z^2} + \rho_B \nu_i v_A$$  \hspace{1cm} (3.13)
The energy equation may be written as:

\[ \rho_g c_{pg} \frac{\partial T}{\partial t} = -u_z \rho_g c_{pg} \frac{\partial T}{\partial z} + \lambda_{eff} \frac{\partial^2 T}{\partial z^2} + (\Delta H) \rho_B v_l r_A - U_{tw}(T - T_w) \]  

(3.14)

The rate of reaction is given as:

\[ r_A = k_0 e^{-E/RT} C_A^n \]  

(Power Law Model)

\[ r_A = k_0 e^{-E/RT} \left( \frac{F_A}{K_P K_{PB}} \right)^{1/2} \left( \frac{F_B}{1 + K_A F_B + K_B F_D} \right)^{1/2} \]  

(Mechanistic Model)

where the parameters are as defined: \( C_A \) is the concentration of the reactant; \( T \) is the average temperature within the reactor; \( T_w \) is the temperature of the heating fluid; \( u_z \) is the superficial velocity of the fluid (e.g. gases) mixture; \( \rho_B \) is the bulk density of the fixed bed; \( \rho_g \) is the gas mixture density; \( r_A \) is the rate of reaction for component A. For this reaction it was negative because the component (methane) was being consumed (reactant); \( c_{pg} \) is the heat capacity of the gas mixture; \( \Delta H \) is the heat of reaction; \( U_{tw} \) is the overall heat transfer coefficient; \( D_{eff} \) is the effective diffusivity in the axial direction; \( \lambda_{eff} \) is the effective axial conductivity; \( n \) is the reaction order; \( v_i \) is the stoichiometric coefficient of component A (in this case methane); \( k_0 \) is the pre-exponential factor; \( E \) is the activation energy; \( R \) is the gas constant; \( F_i \) is the flow rate of the chemical species; \( K_P \) is the thermodynamic equilibrium constant; \( K_A \) is the adsorption constant for component A.

The model was selected after a critical review of the models used in the literature. It was observed that the models applied were an approximation of the proposed model based on the assumptions made in the model development. This model is such that it
can be scaled up to a two dimensional model of the process by including diffusion in both axial and radial direction. This would make the model more comprehensive but at the cost of increasing the complexity. On the other hand, the model can be simplified further by making a few more assumptions about the process. For this research work, the model was used for simulating the dynamics of the plug flow reactor.

3.4 Model Predictive Control (MPC) Theory

Model predictive control (MPC) is the control strategy that was adopted for controlling the temperature within the plug flow reactor. MPC refers to a range of control methods that make an explicit use of a model of the process to obtain the control signal by minimizing an objective function. The various models used in the design procedure accounts for the differences found amongst the various available MPC strategies. These design procedures lead to linear controllers that have practically the same structure, and present adequate degrees of freedom. The objective function minimized is a quadratic function given in equation 3.15 below:

\[
J = \sum_{t=k}^{k+N_p} (w(t) - y(t))^T Q_{ctt} (w(t) - y(t)) + \sum_{t=k}^{k+N_c} (u(t)^T R_{ctt} u(t))
\]  

(3.15)

where \( J \) is the objective function, \( u(t) \) is the input, \( y(t) \) is the output, \( w(t) \) is the reference signal, \( N_p \) is the prediction horizon, \( N_c \) is the control horizon, \( \delta u(t) = u(t) - u(t - 1) \), \( Q_{ctt} \) and \( R_{ctt} \) weighting matrices.

The developments till date have yielded far reaching results. The reason for this success can be attributed to the fact that MPC is, perhaps, the most general way of
posing the process control problem in the time domain. MPC formulations integrate optimal control, stochastic control, control of processes with dead time, multivariable control, and future references when available. Another advantage of model predictive control is that because of the finite control horizon used, constraints, and in general nonlinear processes, which are frequently found in industry, can be handled. On the other hand, although a number of applications have been reported both in industry and research institutions, model predictive control has not yet reached in industry the popularity that its potential would suggest. One of the reasons for this is that its implementation requires some mathematical complexities that are not a problem in general for the research control community, where mathematical packages are normally fully available, but represent a drawback for the use of the technique by control engineers in practice. While MPC may be suitable for almost any kind of problem, it displays its main strength when applied to problems with:

- A large number of manipulated and controlled variables
- Constraints imposed on both the manipulated and controlled variable
- Changing control objectives and/or equipment failure
- Time delays

Some of the more common implementations of MPC are Dynamic Matrix Control (DMC), Model Algorithmic Control (MAC), and Generalized Predictive Control (GPC). While these algorithms differ in certain details, the main ideas behind them are very similar, which are:

- The use of a model to predict the process output at future time instants
• Calculation of a control sequence by minimizing an objective function

• Receding strategy so that at each instant the horizon is displaced towards the future.

In its unconstrained form, MPC is closely related to linear quadratic (LQ) optimal control. When constraints are considered, MPC becomes an optimization problem that is solved online in real-time at each sampling interval. MPC differs from other control methods mainly in its implementation of the control actions. Usually, MPC solves a finite horizon optimal control problem at each sampling instant, so that the control moves for the current time and a period of future time instants are obtained. However, only the current control move is applied to the plant. At the next sampling instant, the same optimization is repeated with the new measurements. There are many applications of predictive control successfully in use at the present time, not only in the process industry, but also applications to the control of a diversity of processes including robotic manipulators and administration of anesthesia. The good performance of these applications shows the capacity of the MPC to achieve highly efficient control systems that can operate during long periods of time with hardly any intervention (Bao-Cang, 2010).
3.4.1 MPC Strategy

Figure 3.2 MPC Strategy (Espinosa et al., 2005)

The methodology of all the controllers belonging to the MPC family are characterized by a set of common elements, which when translated into this research work can be applied as follows:

1. The future outputs for a determined horizon $N_p$, called the prediction horizon, are predicted at each instant $t$ using the process model. These predicted outputs $y(t+k|t)$ for $k = 1,…,N_p$ depend on the known values of inputs and outputs up to instant $t$ and on the future control signals $u(t+k|t)$, $k=0,…,N_c-1$, which are those to be sent to the system and to be calculated.

2. The set of future control signals is calculated by optimizing a determined criterion in order to keep the process as close as possible to the reference trajectory $w(t+k)$. This criterion usually takes the form of a quadratic function
of the errors between the predicted output signal and the predicted reference trajectory. The control effort is included in the objective function in most cases. An explicit solution can be obtained, if the criterion is quadratic, the model is linear, and there are no constraints, otherwise an iterative optimization method has to be used. Some assumptions about the structure of the future control law are also made in some cases, such as that it will be constant from a given instant.

3. The control signal \( u(t|t) \) is sent to the process whilst the next control signal calculated are rejected, because at the next sampling instant \( y(t+1) \) is already known, and step 1 is repeated with this new value, and all the sequences are brought up to date. Thus, the \( u(t+1|t+1) \) is calculated using the receding horizon concept.

According to Espinosa et al. (2005), this can be expressed as an algorithm as shown below:

1. Sample the output of the plant.

2. Use the model of the plant to predict its future behavior over a prediction horizon during \( N_p \) samples when a control action is applied along a control horizon during \( N_c \) samples.

3. Calculate the optimal control sequence \( \{u(k),\ldots,u(k+N_c)\} \) that minimizes

\[
\min_{u(k),\ldots,u(k+N_c)} J(u(k), y(k), w(k))
\]
Subject to

\[
x(k + 1) = f(x(k), u(k))
\]

\[
y(k) = g(x(k), u(k))
\]

\[
y_{\min} \leq y(k) \leq y_{\max}, \forall k = 1, \ldots, N_p
\]

\[
u_{\min} \leq u(k) \leq u_{\max}, \forall k = 1, \ldots, N_c
\]

\[
\Delta y_{\min} \leq \Delta y(k) \leq \Delta y_{\max}, \forall k = 1, \ldots, N_p
\]

\[
|\Delta u(k)| \leq \Delta u_{\max}, \forall k = 1, \ldots, N_c
\]

where \( J(.) \) is the cost function expressed as;

\[
J(u(k), y(k), r(k)) = \sum_{t=k}^{k+N_p} (w(t) - y(t))^T Q_{cte} (w(t) - y(t)) + \sum_{t=k}^{k+N_c} (u(t)^T R_{cte} u(t))
\]

where \( x(k) \) represents the states of the system, \( u(k) \) the inputs, \( y(k) \) the outputs, the functions \( f(.,.) \) and \( g(.,.) \) represents the dynamic model of the plant, \( w(k) \) is the reference signal, \( \delta u(t) = u(t) - u(t-1) \). \( Q_{cte} \) and \( R_{cte} \) are weighting matrices.

4. Apply the input \( u(k) \) and repeat the procedure at the next sampling time.

In order to implement this strategy, a model is used to predict the future plant outputs, based on past and current values on the proposed optimal future control actions. The process model plays, in consequence, a decisive role in the controller. The chosen model must be capable of capturing the process dynamics so as to precisely predict the future outputs as well as being simple to implement and to understand. As MPC is not a unique technique but a set of different methodologies, there are many types of models used in various formulations.
3.5 Nonlinear Predictive Control

Linear Model Predictive Control has over the years become widely accepted as an established research field (Espinosa et al., 2005, Camacho et al., 2010). This involves the control of linear processes with linear constraints. When processes are kept close to an operating point, linear models are good approximations. These models also work in cases where the nonlinearities are not too severe. However, this is not the case in the process industries (Camacho et al., 2010). Quite a number of processes in the chemical process industry are nonlinear to varying degrees of severities. In most cases, these processes operate around steady states. This, in essence, enables the use of a linear representation of the system in the controller design. There are exceptions to this approach; such as in situations where the nonlinearity is so severe, even around the steady states, and so crucial to the closed loop stability that a linear model is not sufficient. Similarly in processes with continuous transitions (start-ups, shut-down, etc.) that spend a lot of time away from steady state regions, or in processes that are never in steady state operations like the batch processes where the whole operation is carried out in transient mode, the use of a linear control law will not be very effective. Controllers based on a nonlinear model will be essential for improved performance or simply for a stable operation.

3.6 Nonlinear system identification

In the area of nonlinear system identification, several schemes could be adopted. Neural networks, Volterra series and Wavelets represent some of the universal approximators used in nonlinear system identification. System identification is a
technique to build mathematical models of dynamic systems based on input – output data. The data set is described as

\[ N^t = \{y(1), u(1), ..., y(t), u(t)\} \]  

(3.16)

where \( y(t) \) is the output of the dynamic system at time \( t \) and \( u(t) \) is the input.

A model of the dynamic system can be constructed as a mapping from past data \( N^{t-1} \) to the next output \( y(t) \). This model is known as the predictor model and is represented by

\[ \hat{y}(t) = f(N^{t-1}) \]  

(3.17)

where \( \hat{y}(t) \) represents the estimated output. The essence of identification using, for instance, fuzzy systems is to try to represent the function \( f \) by means of a fuzzy model. It is important to see the fuzzy system as a parameterizable mapping,

\[ \hat{y}(t|\theta) = f(N^{t-1}|\theta) \]  

(3.18)

where \( \theta \) is the vector of parameters to be chosen, for example the shape of the membership functions. The choice of these parameters is guided by the information embedded in the data. The structure of \( \{3.16\} \) is generic with the drawback that the data set is continuously increasing. For this reason, it is better to use a vector \( \varphi(t) \) of fixed dimension. So the general model is now formulated as

\[ \hat{y}(t|\theta) = f(\varphi(t)|\theta) \]  

(3.19)

The vector \( \varphi \) is known as the regression vector and its elements regressors.
\[ \varphi(t) = [y(t - 1), ..., y(t - n), u(t - 1), ..., u(t - m)] \]  

(3.20)

Using this parameterization, the problem is divided into three sub-problems:

(1) Choice of regressors in \( \varphi(t) \) from the set of past inputs and outputs

(2) Finding the structure of the Fuzzy system \( f(...) \)

(3) How to find the parameters \( \theta \)

### 3.6.1 Nonlinear System Identification using a Fuzzy Model

The advantages of the use of fuzzy systems is their capacity to interact and to extract linguistic information from input – output data, and to describe the dynamics of the system in local regions described by the rules. These features are very valuable and make fuzzy models different from other traditional black-box techniques. The modeling task is carried out by fuzzy inference system (FIS). Fuzzy Inference Systems are processing units that convert numerical information into linguistic variables by means of a fuzzification process, process the linguistic information using a rule base, and generate a numerical result from the conclusions of the rules by means of the defuzzification process. Fuzzy inference systems are universal approximators. This property means that FISs are capable of approximating any continuous function into a compact domain with a certain level of accuracy.

Fuzzy models can be dynamic or static. Different types of fuzzy models have been proposed but the most used are the rule-based fuzzy system. These models are characterized by having fuzzy propositions as antecedents and consequences of the rules (Mamdani models). Another important type of fuzzy model is the Takagi –
Sugeno fuzzy model, in which the consequences of the rules are crisp functions of the antecedents (Espinosa et al., 2005).

3.6.2 Nonlinear System Identification using a Neuro-Fuzzy Model

Neural methods provide learning capability, whereas fuzzy methods provide flexible knowledge-representational capability. Integrating these two methodologies can lead to better technologies that take advantage of the strengths of each methodology, and at the same time overcome some of the limitations of the individual techniques.

There are basically two ways that fuzzy and neural technologies can be combined. In one direction, fuzzy logic can be introduced into neural networks to enhance knowledge representation capability of conventional neural networks. This can be done by introducing fuzzy concepts within neural networks at the levels of inputs, weights, aggregation operations, activation functions, and outputs. Standard mathematical models for neurons can, for example, be changed to “fuzzy-neurons” with t-norms and t-conorms used to build aggregation operations. This leads to a fuzzy-neural system, which one can present fuzzy inputs and develop an analog of the conventional back-propagation algorithm for training.

In another direction, neural networks can be used in fuzzy modeling and control to provide fuzzy systems with learning capabilities. These methods lead to neural-fuzzy system that is a fuzzy system represented as a modified neural network. The resulting fuzzy inference system is enhanced by neural network capabilities. Fuzzy systems are generally more “user friendly” than neural systems because their behavior can be
explained based on fuzzy rules fashioned after human reasoning. Although fuzzy logic can encode expert knowledge directly using rules with linguistic labels, its design and membership function (which represent the linguistic labels) tuning is usually time consuming. Neural network learning techniques can automate this process and substantially reduce development time and cost while improving performance. Neural networks are also used to preprocess data, and to extract fuzzy control rules from numerical data automatically, as well as to tune membership functions of fuzzy systems (Nguyen et al., 2003). An example of a neural-fuzzy system is the adaptive neuro-fuzzy inference system (ANFIS).

### 3.6.2.1 Adaptive Neuro-Fuzzy Inference System (ANFIS) Architecture

ANFIS is functionally equivalent to a Fuzzy Inference Systems. For simplicity we assume that the fuzzy inference system under consideration has two inputs $x$ and $y$ and one output $z$. For a first order Sugeno fuzzy model, a common rule set with two fuzzy “if – then” rules are given by the following:

**Rule 1:** If $x$ is $A_1$ and $y$ is $B_1$, then $f_1 = p_1 x + q_1 y + r_1$.

**Rule 2:** If $x$ is $A_2$ and $y$ is $B_2$, then $f_2 = p_2 x + q_2 y + r_2$. 
Figure 3.3 A two-input first-order Sugeno fuzzy model with two rules (Jang et al., 1997)

Figure 3.4 Equivalent ANFIS architecture (Jang et al., 1997)

Figure 3.3 above illustrates the reasoning mechanism for the Sugeno model. The corresponding equivalent ANFIS architecture is shown in Figure 3.4 above where nodes of the same layer have similar functions, as described below.
Layer 1

Every node $i$ in this layer is an adaptive node with a node function

$$O_{1,i} = \mu_{A_i}(x), \quad \text{for } i = 1, 2,$$

$$O_{1,i} = \mu_{B_{i-2}}(y), \quad \text{for } i = 3, 4,$$

where $x$ (or $y$) is the input to node $i$ and $A_i$ (or $B_{i-2}$) is a linguistic label (such as “small” or “large”) associated with this node. In other words, $O_{1,i}$ is the membership grade of a fuzzy set $A_1 = A_2 = B_1 = B_2$ and it specifies the degree to which the given input $x$ (or $y$) satisfies the quantifier $A$. Here the membership function for $A$ can be any appropriate parameterized membership function, such as the generalized bell function:

$$\mu_A(x) = \frac{1}{1 + \left| \frac{x-c_i}{a_i} \right|^{2b}} \quad (3.21)$$

where \{a_i, b_i, c_i\} is the parameter set. As the values of these parameters change, the bell-shaped function varies accordingly, thereby exhibiting various forms of membership functions for fuzzy set $A$. Parameters in this layer are referred to as premise parameters.

Layer 2

Every node in this layer is a fixed node labeled $\Pi$, whose output is the product of all the incoming signals:

$$O_{1,i} = w_i = \mu_{A_i}(x)\mu_{B_i}(y), \quad \text{for } i = 1, 2.$$
Each node output represents the firing strength of a rule. In general, any other T–norm operators that perform fuzzy AND can be used as the node function in this layer.

**Layer 3**

Every node is this layer is a fixed node labeled N. the \( i \)th node calculates the ratio of the \( i \)th rule’s firing strength to the sum of all rules’ firing strengths:

\[
O_{3,i} = \tilde{w}_i = \frac{w_i}{w_1 + w_2}, \quad i = 1, 2.
\]

For convenience, outputs of this layer are called normalized firing strengths.

**Layer 4**

Every node \( i \) in this layer is an adaptive node with a node function

\[
O_{4,i} = \tilde{w}_i f_i = \tilde{w}_i (f_1 = p_i x + q_i y + r_i)
\]

where \( \tilde{w}_i \) is a normalized firing strength from layer 3 and \( \{p_i, q_i, r_i\} \) is the parameter set of this node. Parameters in this layer are referred to as consequent parameters.

**Layer 5**

The single node in this layer is a fixed node labeled \( \Sigma \), which computes the overall output as the summation of all incoming signals

\[
\text{overall output } = O_{5,1} = \sum_i \tilde{w}_i f_i = \frac{\sum_i w_if_i}{\sum_i w_i}
\]

Thus, we have constructed an adaptive network that is functionally equivalent to a Sugeno fuzzy model.
3.6.2.2 Hybrid Learning Algorithm

From ANFIS architecture shown in Figure 3.4, we observe that when the values of the premise parameters are fixed, the overall output can be expressed as a linear combination of the consequent parameters. In symbols, the output $f$ in Figure 3.4 above can be rewritten as

$$f = \frac{w_1}{w_1+w_2}f_1 + \frac{w_2}{w_1+w_2}f_2$$

$$= \bar{w}_1(p_1x + q_1y + r_1) + \bar{w}_2(p_2x + q_2y + r_2)$$

$$= (\bar{w}_1x)p_1 + (\bar{w}_1y)q_1 + (\bar{w}_1)r_1 + (\bar{w}_2x)p_2 + (\bar{w}_2y)q_2 + (\bar{w}_2)r_2$$

which is linear in the consequent parameters $p_1, q_1, r_1, p_2, q_2, r_2$. From this observation, we have

$$S = \text{set of total parameters}$$

$$S_1 = \text{set of premise (nonlinear) parameters},$$

$$S_2 = \text{set of consequent (linear) parameters}$$

In the forward pass of the hybrid learning algorithm, node outputs go forward until layer 4 and the consequent parameters are identified by the least squares method. In the backward pass, the error signals propagate backward and the premise parameters are updated by gradient descent.

The identified consequent parameters are optimal under the condition that the premise parameters are fixed. Accordingly, the hybrid approach converges much faster since it
reduces the search space dimensions of the original pure back-propagation method. Thus, we should always look for the possibility of decomposing the parameter set in the first place. For Tsukamoto ANFIS, this can be achieved if the membership function on the consequent part of each rule is replaced by a piecewise linear approximation with two consequent parameters, as shown in Figure 3.5 below. In this case, again, the consequent parameters constitute the linear parameter set $S_2$ and the hybrid learning rule can be employed as before.

![Figure 3.5 Piecewise linear approximation of consequent MFs in Tsukamoto ANFIS](Jang et al., 1997)

### 3.7 COMSOL Multiphysics

COMSOL Multiphysics is a powerful and interactive software for modeling and solving all kinds of scientific and engineering problems. With this software you can easily extend conventional models for one type of physics into multiphysics models that solve coupled physics phenomena simultaneously. Using the built-in physics
interfaces and the advanced support for material properties, it is possible to build models by defining the relevant physical quantities—such as material properties, loads, constraints, sources, and fluxes—rather than by defining the underlying equations. One can always apply these variables, expressions, or numbers directly to solid and fluid domains, boundaries, edges, and points independently of the computational mesh. COMSOL Multiphysics then internally compiles a set of equations representing the entire model.

COMSOL Multiphysics offers modeling and analysis power for many application areas. Many real-world applications involve simultaneous couplings in a system of PDEs—multiphysics. For several of the key application areas there are optional modules. The two modules that were utilized in this research were the Chemical Reaction Engineering Module and the Heat Transfer module.

3.7.1 Chemical Reaction Engineering Module

In this module, reaction engineering tools use reaction formulas to create models of reacting systems, where a model means the material (mass), energy (heat), and momentum balances for a system. The Chemical Reaction Engineering Module not only defines these balances, it can also solve the material and energy balances for both space-independent and space-dependent models. This makes it possible to create models involving material, energy, and momentum balances in COMSOL Multiphysics directly from a set of reaction formulas. The interface used in this instance was the Transport of Diluted Species interface (COMSOL Multiphysics user’s guide, 2010).
3.7.2 Transport of Diluted Species Interface

As stated in COMSOL Multiphysics user’s guide (2010), the Transport of Diluted Species interface provides a predefined modeling environment for studying the evolution of chemical species transported by diffusion, convection, and migration due to an electric field. The interface assumes that all species present are dilute, that their concentration is small compared to a solvent fluid or solid. As a rule of thumb, a mixture containing several species can be considered dilute when the concentration of the solvent is more than 90 mol%. Due to the dilution, mixture properties such as density and viscosity can be assumed to correspond to those of the solvent. Fick’s law governs the diffusion of the solutes dilute mixtures or solutions, while the phenomenon of ionic migration is sometimes referred to as electrokinetic flow.

The default node attributed to the Transport of Diluted Species interface assumes chemical species transport through diffusion and implements the mass balance equation:

\[
\frac{\partial c}{\partial t} + \mathbf{u} \cdot \nabla c = \nabla \cdot (D \nabla c) + R 
\]  \hspace{1cm} (3.24)

where:

\( \nabla \) is the gradient function

\( c \) is the concentration of the species (mol/m\(^3\))

\( D \) denotes the diffusion coefficient (m\(^2\)/s)

\( R \) is a reaction rate expression for the species (mol/(m\(^3\)·s))
\( \mathbf{u} \) is the velocity vector (m/s). The first term on the left-hand side of Equation.

### 3.7.3 Heat Transfer Module

The Heat Transfer Module supports all fundamental mechanisms of heat transfer, including conductive, convective, and radiative heat transfer (both surface-to-surface and surface-to-ambient radiation). Using the physics interfaces in this module along with inherent multiphysics capabilities of COMSOL Multiphysics one can model a temperature field in parallel with other physics, which makes models even more accurate and representative of the real world. For this instance, the interface used was the Heat Transfer in Fluids interface (COMSOL Multiphysics user’s guide, 2010).

### 3.7.4 Heat Transfer in Fluids Interface

The Heat Transfer in Fluids feature uses the following version of the *heat equation* as the mathematical model for heat transfer in fluids: with the following material properties:

\[
\rho C_p \frac{\partial T}{\partial t} + \rho C_p \mathbf{u} \cdot \nabla T = \nabla \cdot (k \nabla T) + Q \tag{3.25}
\]

where:

- \( \rho \) is the *density*
- \( C_p \) is the fluid *heat capacity* at constant pressure and it describes the amount of heat energy required to produce a unit temperature change in a unit mass
- \( k \) is the fluid *thermal conductivity* (a scalar or a tensor, if the thermal conductivity is anisotropic)
- \( \mathbf{u} \) is the fluid velocity field, which can be an analytic expression or a velocity field from a fluid-flow interface
$Q$ is the heat source (or sink); one or more heat sources can be added separately.

For a steady-state problem the temperature does not change with time and the first term disappears. The ratio of specific heats $\gamma$ is defined as the ratio of heat capacity at constant pressure, $C_p$, to heat capacity at constant volume, $C_v$. When using the ideal gas law to describe a fluid, specifying $\gamma$ is enough to evaluate $C_p$. For common diatomic gases such as air, $\gamma = 1.4$ is the standard value. Most liquids have $\gamma = 1.1$ while water has $\gamma = 1.0$. $\gamma$ is used in the streamline stabilization and in the postprocessing variables for heat fluxes and total energy fluxes (COMSOL Multiphysics user’s guide, 2010).

Chapter four illustrates how COMSOL Multiphysics was used to design the model for the research as well as to generate data for training ANFIS. The results of the simulations and the accompanying analysis were also presented in this chapter.
4. ANALYSIS AND RESULTS

4.1 Controller Design Strategy

This research work was broken down into several phases. The first phase primarily focused on the steady state simulation of the process. This was done in Comsol multiphysics, and it was necessary to replicate the operating conditions and results obtained by Akpan et al. (2007). Once this was validated, the model was scaled up to account for the transient response of the process. This was done in the second phase, and it enabled the dynamics of the reactor to be captured. Also, in this phase, the data that was used to train the ANFIS model was generated. In the third phase, the ANFIS model was trained using the training and checking data set. Once the training was completed, the model was validated using the validating data set. The final phase involved the implementation of the control algorithm. An MPC control scheme was implemented as the control strategy to regulate the temperature within the reactor. The third and final phases were implemented in Matlab.

4.2 Phase 1 - Steady State Simulation of the Plug Flow Reactor in Comsol Multiphysics

The first phase of this research work involved simulations to replicate the results reported by Akpan et al. (2007) in order to validate the model used. This simulation was carried out in the chemical reaction engineering module of Comsol multiphysics. The parameters used in the simulation are as given in the table below:
<table>
<thead>
<tr>
<th>Parameter</th>
<th>Definition and units</th>
<th>Values</th>
</tr>
</thead>
<tbody>
<tr>
<td>$T_0$</td>
<td>Feed inlet temperature, K</td>
<td>973</td>
</tr>
<tr>
<td>$u_x$</td>
<td>Superficial velocity, km/h</td>
<td>1.3</td>
</tr>
<tr>
<td>$W/A_{A0}$</td>
<td>Space time, kg-cat s/kg-CH₄</td>
<td>$1.6 \times 10^{-2}$</td>
</tr>
<tr>
<td>$D_p$</td>
<td>Catalyst particle diameter, mm</td>
<td>0.3</td>
</tr>
<tr>
<td>$D_t$</td>
<td>Internal diameter of the tube of the reactor, mm</td>
<td>6.3</td>
</tr>
<tr>
<td>$R_t$</td>
<td>Radius of packed bed tubular reactor, m</td>
<td>$3.15 \times 10^{-3}$</td>
</tr>
<tr>
<td>$L$</td>
<td>Catalyst bed length, m</td>
<td>$3 \times 10^{-2}$</td>
</tr>
<tr>
<td>$\rho_g$</td>
<td>Gas density, kg/m³</td>
<td>0.37</td>
</tr>
<tr>
<td>$\rho_B$</td>
<td>Bulk density of catalyst in the reactor, kg/m³</td>
<td>53.5</td>
</tr>
<tr>
<td>$\lambda_x$ and $\lambda_r$</td>
<td>Effective thermal conductivity, kJ/m s K</td>
<td>$1.8 \times 10^{-3}$</td>
</tr>
<tr>
<td>$c_p$</td>
<td>Heat capacity, kJ/kg K</td>
<td>2.1</td>
</tr>
<tr>
<td>$D_x$ and $D_r$</td>
<td>Effective diffusivity, m²/h</td>
<td>$1.2 \times 10^{-3}$</td>
</tr>
<tr>
<td>$\Delta H$</td>
<td>Heat of reaction, kJ/kmol</td>
<td>$2.6 \times 10^5$</td>
</tr>
<tr>
<td>$U_{TW}$</td>
<td>Heat transfer coefficient, kJ/m²h K</td>
<td>$1.8 \times 10^2$</td>
</tr>
<tr>
<td>$k_0$</td>
<td>Collision coefficient</td>
<td>$1.1 \times 10^{19}$</td>
</tr>
<tr>
<td>$E$</td>
<td>Activation energy, kJ/kmol</td>
<td>$2.2 \times 10^5$</td>
</tr>
<tr>
<td>$K_A$</td>
<td>Adsorption constant</td>
<td>27.7</td>
</tr>
<tr>
<td>$P_{tot}$</td>
<td>Total pressure, kPa</td>
<td>101.3</td>
</tr>
<tr>
<td>$K_P$</td>
<td>Equilibrium constant at $T_0$</td>
<td>7.5</td>
</tr>
<tr>
<td>$F_{A0}$</td>
<td>Inlet molar flow rate of CH₄, kmol/h</td>
<td>$2.1 \times 10^{-4}$</td>
</tr>
<tr>
<td>$F_{B0}$</td>
<td>Inlet molar flow rate of CO₂, kmol/h</td>
<td>$2.0 \times 10^{-4}$</td>
</tr>
<tr>
<td>$F_{N0}$</td>
<td>Inlet molar flow rate of N₂, kmol/h</td>
<td>$1.0 \times 10^{-4}$</td>
</tr>
<tr>
<td>$y_A^0$</td>
<td>Inlet mole fraction of CH₄</td>
<td>0.4</td>
</tr>
<tr>
<td>$y_B^0$</td>
<td>Inlet mole fraction of CO₂</td>
<td>0.4</td>
</tr>
<tr>
<td>$y_{N2}^0$</td>
<td>Inlet mole fraction of N₂</td>
<td>0.2</td>
</tr>
<tr>
<td>$M_{ave}$</td>
<td>Average molecular weight of feed mixture</td>
<td>29.5</td>
</tr>
</tbody>
</table>

Table 4.1 Operating conditions and parameters used in the simulation

A 2 dimensional axisymmetric stationary simulation setup in Comsol multiphysics was used to duplicate the results reported by Akpan et al. (2007). In this setup, the reactor was assumed to be symmetrical around the radial axis. For the purposes of this simulation, Transport of Diluted Species (chds) was used for the component balance (equation 3.1) and Heat Transfer in Fluids (ht) was used for the energy balance.
(equation 3.2). A rectangular geometry, with a height of 0.03m and a width of 0.00315m, was used to model the catalyst bed. The operating conditions listed in Table 4.1 above were used during the simulation. In addition, the external heat supply was kept constant at 973 K. The results obtained during this simulation phase are presented below. Figures 4.1 to 4.3 represent the surface plots for methane concentration, methane conversion and temperature within the reactor. For the surface plots, the x-axis is the radius of the reactor from the center to the wall whilst the y-axis represents the length of the reactor. It can be observed that the majority of the reaction occurs early in the reactor.

Figure 4.1 Surface plot of concentration
The total volumetric flow rate was calculated from the total molar flow rate using the equation below.

\[ V = \frac{F_{tot} \times R \times T_0}{P_{tot}} \]  \hspace{1cm} (4.1)

where \( V \) is the total volumetric flow rate of the feed; \( F_{tot} \) is the total molar flow rate of the feed; \( R \) is the gas constant; \( T_0 \) is the feed inlet temperature; \( P_{tot} \) is the total pressure. The total volumetric flow rate was used to calculate the inlet concentration of the reacting species according to the formula in equation 4.2 below.

\[ c_{i0} = \frac{F_{i0}}{V} \]  \hspace{1cm} (4.2)

where \( c_{i0} \) is the initial concentration of species \( i \); \( F_{i0} \) is the inlet molar flow rate of species \( i \); \( V \) is the total volumetric flow rate of the feed. For the results presented methane was used.
Methane conversion was calculated according to the formula given in equation 4.3 below:

\[
\text{Conversion (CH}_4\text{)} = \frac{(CH_4)_\text{in} - (CH_4)_\text{out}}{(CH_4)_\text{in}}
\]  

(4.3)

where \((CH_4)_\text{in}\) is the concentration of the methane feed; \((CH_4)_\text{out}\) is the concentration of methane measured at the outlet of the reactor.
To validate the model used for this simulation, the conversion obtained was compared to what was reported by Akpan et al. (2007) at $W/F_{A0} = 0.016h$ where $W$ is the weight of catalyst and $F_{A0}$ is the feed molar flow rate of methane. This is shown in Figure 4.4 below.
Figure 4.4 Methane conversion profile in the axial direction

The methane conversion profile reported by Akpan et al. (2007) had a maximum value of about 70% at the end of the catalyst bed, and this was comparable to what was obtained in this simulation. This result is shown in Figure 4.4 above, and it confirms that the feed conversion slows down as it travels down the catalyst bed as a result of the reduction in the concentration of the reactant species. The variance observed between the two plots was due to the rate equation used in the simulation. In this case the power law model was used as compared to the mechanistic model used by Akpan et al in their study.
4.3 Phase 2 - Transient Response of the Plug Flow Reactor

The transient response of the reactor was necessary for the design of an appropriate controller for the process. This study reports on how the reactor performs over time. The mathematical model introduced in chapter 3 (equations 3.13 and 3.14) was used in the Comsol simulation for this study. The measurements were taken at the center of the reactor outlet.

Figure 4.5 Transient response of the reactor (Conversion profile)
In Figure 4.5 above, it can be observed that the maximum conversion is about 70%. This value is comparable to what was obtained at the end of the reactor for the steady state simulation. The observed overshoot is a result of the system being a second order system. An assumption made during the model modification was that the radial variations in the parameters were negligible. The plot shown in Figure 4.6 below validates this assumption.

Figure 4.6 Transient response of the reactor measured at 3 points at the reactor outlet
The basis for the validation is the fact that the values for the conversion are very close together. For this test, measurements were taken at 3 different positions at the reactor outlet. At the reactor wall, the conversion was the highest because this was the point of application of heat into the reactor. Another observation made was that the system attained steady state after about 0.25 seconds. This is shown in Figure 4.7 below.

Figure 4.7 Transient response of the reactor showing how long it takes to get to steady state.
This test was used to select a sampling time for the process. Figures 4.8 and 4.9 show the transient response of the reactor for the concentration and temperature profiles.

Figure 4.8 Transient response of the reactor (Concentration profile)
Figure 4.9 Transient response of the reactor (Temperature profile)

From Figures 4.8 and 4.9 it can be observed that the values of the concentration and temperature when the process is at rest is similar to the minimum value of concentration and the maximum value of temperature obtained in phase 1.
4.4 Phase 2 - Response of Reactor to Step Change in External Temperature

The step response of a system at any given initial state consists of the time evolution of its outputs when its control input changes from 0 to 1 suddenly. This test was used to study the time response of the reactor to the control input. Figure 4.10 below shows the external temperature that was used to excite the reactor.

![Graph showing step change in external temperature](image)

Figure 4.10 Step change in external temperature

The temperature of the heat input into the reactor was changed from 487 K to 973 K at the 2.5 seconds mark. The response of the reactor to this sudden change in external temperature is shown in Figures 4.11 to 4.13 below:
Figure 4.11 Response of reactor to step change in external temperature (conversion profile)

From Figure 4.11 above it is observed that at a temperature of 487 K, the conversion is about 42%. After a step change in temperature to 973 K at 2.5 seconds, the conversion jumps to 70% thus, explaining the two-step phenomenon shown in Figure 4.11 above. This two-step phenomenon is also observed in the concentration and temperature profiles shown in Figures 4.12 and 4.13 below. The value of the concentration of methane dropped by about 50% after the step input was applied.
Figure 4.12 Response of reactor to step change in external temperature (concentration profile)
As can be seen in the graphs above, the system has a fast response time. Steady state condition was achieved in about 0.3 seconds. It could be said that the reaction that occurs within the reactor is instantaneous.

4.5 Phase 2 - The flow rate term \( (V_w) \)

The model used in simulating the dynamics of the reactor had a limitation. It lacked a control element. The earlier simulations showed how an external temperature input into the system could be manipulated but lacked a control element. In order to
manipulate the temperature within the reactor to maintain the required conversion, the mathematical model had to be modified to include a flow rate term ($V_w$). The defining equation for this is as given in equations 4.4 and 4.5 below.

\[
\frac{∂c_A}{∂t} = -u_z \frac{∂c_A}{∂z} + D_{eff} \frac{∂^2c_A}{∂z^2} + \rho_B v_i r_A \\
\rho_g c_pg \frac{∂T}{∂t} = -u_z \rho_g c_pg \frac{∂T}{∂z} + \lambda_{eff} \frac{∂^2T}{∂z^2} + (ΔH)ρ_B v_i r_A + ̇q
\]

(4.4) (4.5)

where;

\[r_A = k_0 e^{-E/RT} c_A^n\]

\[ ̇q = (V_w ρ_m c_{pm} \left(1 - e^{-U_{tw, A}/V_w ρ_m c_{pm}}\right)(T_w - T))/A\]

$r_A$ is the rate of reaction. For this parameter, the power law model was used, and it was negative because the component (methane) was being consumed. $\dot{q}$ is the total heat flux into the system. This is the heat that was introduced into the reactor by the combustion of methane. The heating was carried out in an outer jacket, and $V_w$ represents the rate which this heat flows in the outer jacket. By introducing $V_w$ into the system, a control element was introduced. The temperature within the reactor can be controlled by manipulating the flow rate of the heat in the outer jacket.

The other parameters are as defined: $c_A$ is the concentration of the reactant; $T$ is the average temperature within the reactor; $T_w$ is the temperature of the heating fluid; $u_z$ is the superficial velocity of the fluid (e.g. gases) mixture; $\rho_B$ is the bulk density of
the fixed bed; $\rho_g$ is the gas mixture density; $\rho_m$ is the density of the fluid in the heat exchanger; $r_A$ is the rate of reaction. For this reaction it was negative because the component was being consumed; $c_{pg}$ is the heat capacity of the gas mixture; $c_{pm}$ is the heat capacity of the methane being consumed; $\Delta H$ is the heat of reaction; $U_{tw}$ is the overall heat transfer coefficient; $D_{eff}$ is the effective diffusivity in the axial direction; $\lambda_{eff}$ is the effective axial conductivity; $n$ is the reaction order; $v_i$ is the stoichiometric coefficient of component A (in this case methane); $k_0$ is the pre-exponential factor; $E$ is the activation energy; $R$ is the gas constant.

Equation 4.5 could be rewritten as shown below in 4.6.

$$\frac{\partial \tau}{\partial t} = -u_z \frac{\partial \tau}{\partial z} + k_1 \frac{\partial^2 \tau}{\partial z^2} + k_2 r_A + k_3$$

(4.6)

where

$$k_1 = \frac{\lambda_{eff}}{\rho_g c_{pg}}$$

$$k_2 = \frac{(\Delta H) \rho_B v_i}{\rho_g c_{pg}}$$

$$k_3 = \frac{\dot{q}}{\rho_g c_{pg}}$$

The source of heat for the reactor was from the combustion of methane in air according to the equation below.

$$CH_4 + 2O_2 \rightarrow CO_2 + 2H_2O + Heat$$

(4.7)
With the introduction of the flow rate of the heating fluid in the mathematical model, the first step was to get a flow value that would give at least 70% conversion at a temperature of 973 K. This value was calculated as a multiple of the volumetric flow rate within the reactor. The volumetric flow rate was calculated from equation 4.1 given above.

The various values for the flow rate tested in the simulation are presented in the Figure 4.14 below.

![Conversion for various flow rates](image)

Figure 4.14 Conversion for various flow rates
From Figure 4.14 above, it can be observed that a conversion of 70% was obtained when the flow rate of the heating fluid was as low as 0.5 cubic millimeters per second. The choice of a flow rate value was based on the amount of energy expended. In an industrial set up the flow rate value with the least amount of energy expended would be selected.

A step change in the flow rate of the heating fluid was simulated at a temperature of 973 K. The temperature in the heat exchanger was kept constant while the flow rate of the heating fluid was varied to study the response of the reactor to these changes. This test involved the step change in the flow rate of the fluid in the heat exchanger. The result of this simulation is shown in the Figures below.
Figure 4.15 Step change in flow rate in outer jacket
Figure 4.16 Response of reactor to step change in flow rate (conversion profile)
Figure 4.17 Response of reactor to step change in flow rate (concentration profile)
To improve the methane conversion, the temperature within the outer jacket was increased to 1073 K. This is an acceptable temperature in an industrial application of this process.

Figure 4.18 Response of reactor to step change in flow rate (temperature profile)
At this temperature we see that the conversion increases from 70% (Figure 4.5) to 82%. This increase would be equivalent to a 17% increase in production.
4.6 Phase 3 - Data for ANFIS Training

Once all the parameters of the modified equations were tested and validated, the simulator (Comsol Multiphysics) was used to generate the training data set for the ANFIS model. This simulation was run for 100 seconds and the data was sampled every 0.01 seconds. The sample rate was necessary because of the instantaneous nature of the reaction. We can recall that the reaction occurred in about 0.25 seconds (Figure 4.7). The simulation was run for 100 seconds in order to obtain adequate data points for ANFIS training. This model was used in designing the control strategy for the reactor. Below are the plots showing the data points that were used for training ANFIS. For this plots the simulation was run for 5 seconds.
Figure 4.20 Random change in flow rate
Figure 4.21 Reactor response to random change in flow rate (conversion profile)
Figure 4.22 Reactor response to random change in flow rate (concentration profile)
Figure 4.23 Reactor response to random change in flow rate (temperature profile)

The random nature of the flow rate of the heating fluid was used in order to guarantee sufficient excitation around the operating point of the reactor. These excitations were such that if it were applied industrially, the saturation limits of the actuators were respected.
4.7 Phase 3 - ANFIS Model and Training

The ANFIS model had a nonlinear autoregressive exogenous (NARX) model structure. A NARX model has the vector of the regressors composed only of past inputs and outputs. The elements of this regressor were selected after a series of tests.
Below are the results from these tests:

<table>
<thead>
<tr>
<th>Membership Function</th>
<th>Model Input</th>
<th>Training Data Error (x10^-5)</th>
<th>Checking Data Error (x10^-5)</th>
<th>Prediction Error (max)</th>
<th>Prediction Error (min)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Triangular MF</td>
<td>X(k), Vw(k), Vw(k-1), Vw(k-2)</td>
<td>1.6058</td>
<td>1.9476</td>
<td>0.0073</td>
<td>-0.0033</td>
</tr>
<tr>
<td>Gaussian MF</td>
<td>X(k), Vw(k), Vw(k-1), Vw(k-2)</td>
<td>1.6021</td>
<td>1.9352</td>
<td>0.0063</td>
<td>-0.0042</td>
</tr>
<tr>
<td>Generalized Bell MF</td>
<td>X(k), Vw(k), Vw(k-1), Vw(k-2)</td>
<td>1.6044</td>
<td>1.9441</td>
<td>0.0072</td>
<td>-0.0048</td>
</tr>
<tr>
<td>Triangular MF</td>
<td>X(k), X(k-1), Vw(k), Vw(k-1)</td>
<td>0.8876</td>
<td>1.5217</td>
<td>0.0044</td>
<td>-0.0046</td>
</tr>
<tr>
<td>Gaussian MF</td>
<td>X(k), X(k-1), Vw(k), Vw(k-1)</td>
<td>0.8665</td>
<td>1.8638</td>
<td>0.0045</td>
<td>-0.0046</td>
</tr>
<tr>
<td>Generalized Bell MF</td>
<td>X(k), X(k-1), Vw(k), Vw(k-1)</td>
<td>0.8616</td>
<td>1.1826</td>
<td>0.0046</td>
<td>-0.0044</td>
</tr>
<tr>
<td>Triangular MF</td>
<td>X(k), X(k-1), X(k-2), Vw(k)</td>
<td>0.7836</td>
<td>7.0957</td>
<td>0.0045</td>
<td>-0.005</td>
</tr>
<tr>
<td>Gaussian MF</td>
<td>X(k), X(k-1), X(k-2), Vw(k)</td>
<td>0.7772</td>
<td>4.2431</td>
<td>0.0044</td>
<td>-0.0047</td>
</tr>
<tr>
<td>Generalized Bell MF</td>
<td>X(k), X(k-1), X(k-2), Vw(k)</td>
<td>0.7774</td>
<td>2.8739</td>
<td>0.0043</td>
<td>-0.0046</td>
</tr>
<tr>
<td>Triangular MF</td>
<td>X(k), X(k-1), X(k-2), Vw(k)</td>
<td>0.7772</td>
<td>3.6206</td>
<td>0.0045</td>
<td>-0.0046</td>
</tr>
<tr>
<td>Gaussian MF</td>
<td>X(k), X(k-1), X(k-2), Vw(k)</td>
<td>0.7762</td>
<td>5.0933</td>
<td>0.0046</td>
<td>-0.0047</td>
</tr>
<tr>
<td>Generalized Bell MF</td>
<td>X(k), X(k-1), X(k-2), Vw(k)</td>
<td>0.7772</td>
<td>3.6208</td>
<td>0.0045</td>
<td>-0.0046</td>
</tr>
<tr>
<td>Triangular MF</td>
<td>X(k), X(k-1), X(k-2), Vw(k-1), Vw(k-2)</td>
<td>0.7823</td>
<td>6.7675</td>
<td>0.0043</td>
<td>-0.005</td>
</tr>
<tr>
<td>Gaussian MF</td>
<td>X(k), X(k-1), X(k-2), Vw(k-1), Vw(k-2)</td>
<td>0.7773</td>
<td>3.7051</td>
<td>0.0046</td>
<td>-0.0049</td>
</tr>
<tr>
<td>Generalized Bell MF</td>
<td>X(k), X(k-1), X(k-2), Vw(k-1), Vw(k-2)</td>
<td>0.7772</td>
<td>3.2798</td>
<td>0.0046</td>
<td>-0.0048</td>
</tr>
<tr>
<td>Triangular MF</td>
<td>X(k), X(k-1), X(k-2), Vw(k), Vw(k-1)</td>
<td>0.7739</td>
<td>0.0113</td>
<td>0.0045</td>
<td>-0.0051</td>
</tr>
<tr>
<td>Gaussian MF</td>
<td>X(k), X(k-1), X(k-2), Vw(k), Vw(k-1)</td>
<td>0.7619</td>
<td>5.5535</td>
<td>0.0044</td>
<td>-0.0051</td>
</tr>
<tr>
<td>Generalized Bell MF</td>
<td>X(k), X(k-1), X(k-2), Vw(k), Vw(k)</td>
<td>0.7609</td>
<td>4.6754</td>
<td>0.0043</td>
<td>-0.0046</td>
</tr>
<tr>
<td>Triangular MF</td>
<td>X(k), X(k-1), X(k-2), Vw(k-1), Vw(k-2)</td>
<td>0.7705</td>
<td>6.2104</td>
<td>0.0044</td>
<td>-0.0051</td>
</tr>
<tr>
<td>Gaussian MF</td>
<td>X(k), X(k-1), X(k-2), Vw(k-1), Vw(k-2)</td>
<td>0.7606</td>
<td>4.8087</td>
<td>0.0044</td>
<td>-0.0056</td>
</tr>
</tbody>
</table>

Table 4.2 Selection of inputs for the ANFIS Model
These tests were limited to either 4 or 5 inputs for the ANFIS model. It can be observed that the error properties of the various inputs tested were similar. Thus, any of those inputs could be used for the ANFIS model. In all the cases considered, the output to ANFIS was $X(k + 1)$. This is shown in table 4.4 below.
<table>
<thead>
<tr>
<th>Membership Function</th>
<th>Model Input</th>
<th>Model Output</th>
<th># of Rules</th>
<th>Simulation time (secs)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Triangular MF</td>
<td>X(k), Vw(k), Vw(k-1), Vw(k-2)</td>
<td>X(k+1)</td>
<td>81</td>
<td>438.204024</td>
</tr>
<tr>
<td>Gaussian MF</td>
<td>X(k), Vw(k), Vw(k-1), Vw(k-2)</td>
<td>X(k+1)</td>
<td>81</td>
<td>449.088384</td>
</tr>
<tr>
<td>Generalized Bell MF</td>
<td>X(k), Vw(k), Vw(k-1), Vw(k-2)</td>
<td>X(k+1)</td>
<td>81</td>
<td>419.309729</td>
</tr>
<tr>
<td>Triangular MF</td>
<td>X(k), X(k-1), Vw(k), Vw(k-1)</td>
<td>X(k+1)</td>
<td>81</td>
<td>483.53227</td>
</tr>
<tr>
<td>Gaussian MF</td>
<td>X(k), X(k-1), Vw(k), Vw(k-1)</td>
<td>X(k+1)</td>
<td>81</td>
<td>435.817919</td>
</tr>
<tr>
<td>Generalized Bell MF</td>
<td>X(k), X(k-1), Vw(k), Vw(k-1)</td>
<td>X(k+1)</td>
<td>81</td>
<td>416.007647</td>
</tr>
<tr>
<td>Triangular MF</td>
<td>X(k), X(k-1), X(k-2), Vw(k)</td>
<td>X(k+1)</td>
<td>81</td>
<td>435.394518</td>
</tr>
<tr>
<td>Gaussian MF</td>
<td>X(k), X(k-1), X(k-2), Vw(k)</td>
<td>X(k+1)</td>
<td>81</td>
<td>487.577733</td>
</tr>
<tr>
<td>Generalized Bell MF</td>
<td>X(k), X(k-1), X(k-2), Vw(k)</td>
<td>X(k+1)</td>
<td>81</td>
<td>447.733039</td>
</tr>
<tr>
<td>Triangular MF</td>
<td>X(k), X(k-1), X(k-2), Vw(k)</td>
<td>X(k+1)</td>
<td>81</td>
<td>445.102541</td>
</tr>
<tr>
<td>Gaussian MF</td>
<td>X(k), X(k-1), X(k-2), Vw(k)</td>
<td>X(k+1)</td>
<td>81</td>
<td>484.134896</td>
</tr>
<tr>
<td>Generalized Bell MF</td>
<td>X(k), X(k-1), X(k-2), Vw(k)</td>
<td>X(k+1)</td>
<td>81</td>
<td>443.047281</td>
</tr>
<tr>
<td>Triangular MF</td>
<td>X(k), X(k-1), X(k-2), Vw(k)</td>
<td>X(k+1)</td>
<td>81</td>
<td>447.711761</td>
</tr>
<tr>
<td>Gaussian MF</td>
<td>X(k), X(k-1), X(k-2), Vw(k)</td>
<td>X(k+1)</td>
<td>81</td>
<td>471.397906</td>
</tr>
<tr>
<td>Generalized Bell MF</td>
<td>X(k), X(k-1), X(k-2), Vw(k)</td>
<td>X(k+1)</td>
<td>81</td>
<td>578.290085</td>
</tr>
<tr>
<td>Triangular MF</td>
<td>X(k), X(k-1), X(k-2), Vw(k), Vw(k-1)</td>
<td>X(k+1)</td>
<td>243</td>
<td>1823.034951</td>
</tr>
<tr>
<td>Gaussian MF</td>
<td>X(k), X(k-1), X(k-2), Vw(k), Vw(k-1)</td>
<td>X(k+1)</td>
<td>243</td>
<td>1846.54759</td>
</tr>
<tr>
<td>Generalized Bell MF</td>
<td>X(k), X(k-1), X(k-2), Vw(k), Vw(k-1)</td>
<td>X(k+1)</td>
<td>243</td>
<td>1611.495212</td>
</tr>
<tr>
<td>Triangular MF</td>
<td>X(k), X(k-1), X(k-2), Vw(k-1), Vw(k-2)</td>
<td>X(k+1)</td>
<td>243</td>
<td>1822.676095</td>
</tr>
<tr>
<td>Gaussian MF</td>
<td>X(k), X(k-1), X(k-2), Vw(k-1), Vw(k-2)</td>
<td>X(k+1)</td>
<td>243</td>
<td>2027.655313</td>
</tr>
</tbody>
</table>

Table 4.3 Number of rules for the models considered

The processing times was dependent on the number of processes running simultaneously on the computing system. That notwithstanding, it was observed that
the number of rules and the processing time increased significantly once the number of inputs was increased. Thus, the models with 5 inputs were eliminated. The inputs to the ANFIS model were \( x(k) = [X(k)X(k - 1)X(k - 2)V_w(k - 1)]^T \) and the output was \( y(k) = X(k + 1) \).

where \( X(k) \) is the conversion and \( V_w(k) \) is the flow rate of the fluid in the outer jacket.

Each input had 3 generalized bell membership functions. The number of rules extracted was \( 3 \times 3 \times 3 \times 3 = 81 \) rules. From simulations carried out in Comsol, 10,000 data points were generated for training ANFIS. This data set was divided into 7,000 data points for the training and checking data set, and 3,000 for the validating data set. From the validating data set, 1,000 data points were used to validate the ANFIS model. The result of this training process is shown in the Figures below.
Figure 4.24 Validation results for the reactor

The y-axis for figure 4.23a was conversion while the y-axis in figure 4.23b was the prediction error. The training had an epoch number of 100, i.e. the training cycle was repeated 100 times. This was to ensure a thorough learning process. At the end of the training, the reported number of linear parameters was 405 while the nonlinear parameters were 36. From Figure 4.23, it can be observed that the quality of the
ANFIS model was very good, and the two signals appear overlapped. Moreover the prediction error was very small, with a maximum of about 0.0049. This model was used in the design of the control strategy for the reactor. It must be stated at this point that the flow rate values used to train the ANFIS model was modified in order for it to be useable by Matlab. These values were divided by $10^{-5}$ before they were used for the training. This modification was accounted for in the control strategy implementation.

The membership function chosen for this work was the “gbell” membership function. This membership function was selected because it had the least number of parameters that would give the least errors after training. Figure 4.24 below shows the membership functions of the ANFIS model before training while figure 4.25 shows the same membership functions after training was carried out.
Figure 4.25 Membership function of the neuro-fuzzy model of the reactor before training
Figure 4.26 Membership function of the neuro-fuzzy model of the reactor after training
In training an ANFIS model, a good model is achieved when the root mean squared error (RMSE) converges. In Figure 4.26 above, it is seen that the error converges after about 70 epochs.

Figure 4.27 Error plots for the training process
4.8 Phase 4 – Control Strategy Implementation

The MPC strategy had methane conversion as the measured variable while the flow rate of the heating fluid in the outer jacket was the manipulated variable. This control procedure was implemented in the following steps:

(1) **Sample the output of the plant:** For this step conversion values that were generated for training ANFIS was used. These values were assumed to be the initial response of the system.

(2) **Use the model of the plant to predict future behavior of the plant:** The predicted future output of the plant was given by the equation below

\[ y(t + k|t) = y_{\text{forced}}(t + k|t) + y_{\text{free}}(t + k|t) \]  \hspace{1cm} (4.8)

where \( y_{\text{forced}}(t + k|t) \) was dependent only on the future increments on the inputs and \( y_{\text{free}}(t + k|t) \) was dependent only on the past inputs and outputs.

\( y_{\text{free}}(t + k|t) \) was given by equation 4.9 shown below

\[ y_{\text{free}}(t + k|t) = f(\hat{y}(t), \hat{y}(t - 1), \hat{y}(t - 2), u(t - 1)) \]  \hspace{1cm} (4.9)

where \( y \) is the output (conversion) and \( u \) the input (flow rate). The hat sign over the \( y \) indicates that these values were estimated. The ANFIS model was used to carry out this estimation. \( u(t) = u(t + 1) = \cdots = u(t + k - 1) = u(t - 1), \) meaning the system was simulated assuming all the future inputs constant and equal to the last input value applied to the plant.

\( y_{\text{forced}}(t + k|t) \) was given by equation 4.10 shown below

\[ y_{\text{forced}}(t + k|t) = \sum_{i=0}^{k-1} g_i \delta u(t + k - i - 1|t) \]  \hspace{1cm} (4.10)
where $g_t$ are the step response coefficients of the plant, calculated on the present operating point by simulating the step response on the model.

(3) **Calculate the optimal control sequence:** The cost function that was minimized in order to calculate the optimal control sequence was given by equation

$$J = (W - Y)^T Q_{ctt} (W - Y) + \Delta U^T R_{ctt} \Delta U$$  \hspace{1cm} (4.11)

where $W$ is the reference vector; $Q_{ctt}$ and $R_{ctt}$ are positive matrices, which were used as weights; $\Delta U$ was the change in the control signal. The predictor, $Y$, was given by equation 4.12 below:

$$Y = G \Delta U + Y_{free}$$  \hspace{1cm} (4.12)

The minimization of the cost function, $J$, was obtained by calculating the input sequence $\Delta U$ such that $\partial J / \partial \Delta U = 0$. The optimal sequence for $\Delta U$ was given as:

$$\Delta U = (G^T Q_{ctt} G + R_{ctt})^{-1} G^T Q_{ctt} (W - Y_{free})$$  \hspace{1cm} (4.13)

(4) **Apply the input to the plant and repeat the procedure:** The input that was applied to the plant at the current time, $t$, is as given below:

$$u(t) = u(t - 1) + \delta u(t)$$  \hspace{1cm} (4.14)

where $\delta u(t)$ is the first element of the vector $\Delta U$.

The step response was estimated using the formula given in equation 4.15 below.

$$g(k - 1) = \frac{y_{step}(t + k|t) - y_{free}(t + k|t)}{du(t)}$$  \hspace{1cm} (4.15)

where $du(t)$ was the step size.
\begin{equation}
\begin{aligned}
y_{step}(t + k | t) &= f(\hat{y}(t), \hat{y}(t-1), \hat{y}(t-2), u(t-1)) \\
\end{aligned}
\end{equation}

With: \( \hat{u}(k) = \begin{cases} 
  u(k) & \forall k \leq t - 1 \\
  u(t - 1) + du(t) & \forall k > t - 1 
\end{cases} \)

The step size \( du(t) \) was chosen such that the value of \( u(t-1)+du(t) \) did not saturate the actuators and it was very close to the predicted value of \( \Delta u(t) \).

The predictive controller was implemented using the following parameters: prediction horizon \( N_p = 4 \), control horizon \( N_c = 3 \), cost matrices \( Q_{ctl} = I_{N_p} \), \( R_{ctl} = 10I_{N_c} \), where \( I^n \) is an identity matrix. The simulation was run for 120 sample times. Figure 4.27 shows the methane conversion obtained when the reference set point was 0.77. The process was observed to overshoot before attaining steady state after about 25 seconds. The overshoot confirms the theory for second order systems.
Figure 4.28 Output response to set point change
From Figure 4.28 above it can be seen that the control signal is very stable.
Deltau is the change in the control signal (flow rate of the heating fluid), and at steady state, deltau is 0. From Figure 4.29 above, it can be observed that the value of deltau was 0 after about 20 seconds. This confirms that the control strategy was able to maintain the system at a steady state after running for 20 seconds.

Figure 4.30 below shows the response of the system with a “stair” reference. The set point for this test was increased from 0.30 to 0.50 in steps of 0.05. Each step in this
stair lasted for a duration of 30 seconds. This test was used to simulate a situation where the process was run at below its optimal capacity.

Figure 4.31 Response of system to “stair” reference
The plots in Figure 4.30 above show that the control strategy adequately maintains the process at the specified reference set point with little or no overshoot in the system. Another test conducted involved applying perturbations to the process at the middle of each step. The magnitude of the applied perturbation was 0.05 as shown in Figure 4.31 below.

![Plot of Applied Perturbation](image)

**Figure 4.32 Applied perturbations**

Figure 4.32 below show the response of the system to these perturbations.
Figure 4.33 Response of system to applied perturbations

This test was used to model a situation where there was an occasional disturbance in the system. From the plots shown in Figure 4.32 above, it can be seen that the system
responded by returning to steady state after the perturbations were removed. It is significant to note that the execution time of the system is an important parameter in evaluating the applicability of this control strategy. The average time for each simulation was about 18 seconds, thereby demonstrating the industrial applicability of this control strategy.

From these results, it can be deduced that the designed control strategy was very stable within the region of operation. This strengthens the argument for the implementation of MPC in the process industry. Another observation made was that the implemented control strategy was able to maintain the required conversion as demonstrated by Figure 4.26, reinforcing that the objective of this research was successfully met.
5. LIMITATIONS, CONCLUSION AND RECOMMENDATION

The purpose of this study was to design and implement a nonlinear model predictive control (NMPC) strategy for the carbon dioxide reforming of methane (CDRM). This strategy modeled a “real world” environment as would be experienced in the process industries. Unlike the work of Akpan et al., which was the foundation of this research, a flow term was introduced that served as a control element. Also the mathematical model of the reactor was modified to include time dependency. Specifically for this work, an Adaptive Neuro-Fuzzy Inference System (ANFIS) model of the plant was used in the design of the controller. The study just like any other had some limitations as outlined below.

5.1 Limitations

(1) The model of the reactor that was designed and used for the purpose of generating data in this study had to be modified to be one dimensional as opposed to being two dimensional. Although a two dimensional model would have been a better replica of a real world scenario it was outside the scope of this work.

(2) In deriving the mathematical model of the plug flow reactor, the fluid velocity is assumed to be a function of the axial position in the tube. However, in this work, the fluid velocity was assumed to be a constant. This limited the flexibility of the model.

(3) Another limitation to the flexibility of the model was most of the parameters defining the model that were temperature dependent was assumed to be constant.
Despite the fact that these assumptions are valid, the model does not exactly duplicate the effect of “real world” scenarios, which would have some temperature dependency. These assumptions were made to reduce computing time and the complexity of the model.

(4) The range of data that was used for exciting the reactor was limited. This placed some restriction on the operating region of the reactor.

(5) Also, the processing times reported in table 4.4 were dependent on the number of processes running simultaneously on the computing system. For a more dedicated system, these processing times may improve significantly.

Irrespective of these limitations, the results obtained for this research were accurate as were reported in Chapter 4. Section 5.2 below summarizes the results obtained in this study and the conclusions derived from them.

5.2 Conclusions

(1) The nonlinear model predictive control strategy was successfully designed and implemented for the CDRM process. This control scheme was based on a neuro-fuzzy model that was trained from data generated in Comsol multiphysics. The validated model was very accurate and suitable for this model-based control strategy (Figure 4.24). This model was very suitable for carrying out predictions, and also for the control design.

(2) The designed MPC strategy was able to bring the process to its reference setpoint and maintain it there for the duration of the simulation. In addition, the controller could work for a diverse range of reference setpoint.
(3) The controller was very robust. It was able to bring the process back to its reference setpoint once the disturbance in the process was removed.

(4) The implemented controller was observed to be very stable.

5.3 Recommendations for future studies

The following suggestions are possible directions that can be pursued to enhance the quality of results obtained to closely mimic actual scenarios:

(1) A two dimensional model of the process can be utilized. This would increase computation significantly, but it would be a better replica for the real world scenario.

(2) The proposed model used in this study was permissible to making some temperature dependent parameters constant in order to reduce complexities. Any further work on this study should consider eliminating the constants to simulate industrial settings.

(3) The fluid velocity can also be computed as a function of the axial position in the tube. This would be more suggestive of everyday situations in industries, in which the flow in a packed bed reactor deviates from the ideal pattern because of variations in flow velocity and mixing effects due to the presence of packing.

(4) Other nonlinear modeling schemes could be studied and the results compared to evaluate how accurately they model the reactor.
REFERENCES


123


