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Intensification of Catalytic Process in Premixed Lean Hydrogen/air Combustion

by

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B.Sc. in Mechanical Engineering, BUET, Bangladesh, 2016 M.Sc. in Mechanical Engineering, BUET, Bangladesh, 2018

A THESIS SUBMITTED IN FULFILMENT OF THE REQUIREMENTS FOR THE DEGREE OF DOCTOR OF PHILOSOPHY

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Abstract

The global energy use is moving towards hydrogen energy to make the energy sector clean, efficient and sustainable. Therefore, ambitious policies globally on hydrogen energy are made and aimed at transitioning away from fossil fuels to meet the goals of climate change through the Paris Climate Agreement. For this, moving to hydrogen fuel use is a feasible and promising solution, especially, to replacing the fossil fuel use in combustion systems. As NO_x emissions pose a major challenge, further innovation and research are crucial for developing low/ultra-low NO_x hydrogen combustion systems. In this regard, catalytic aided combustion widely investigated in the literature, is useful for efficient hydrogen combustion with reducing significant NO_x emission at low temperatures. However, given the limitations of noble metal use and its high cost, fundamental research is required to optimise the catalytic reactor design with minimal use of expensive catalysts.

The present study begins by numerically investigating the premixed combustion of H₂/air over a platinum catalyst in a planar monolithic reactor (a block with parallel channels resembling a honeycomb structure), with the goal of stabilising the flame under lean to ultralean conditions. A steady laminar species transport model is initially used, incorporating elementary heterogeneous and homogeneous chemical reaction schemes, and the results are validated against experimental data. A stability map for the equivalence ratios (φ) of 0.15 to 0.20 is obtained from a non-catalytic burner, forming the basis for the catalytic flame analysis. In the non-catalytic burner, no flame is observed for $\varphi \leq 0.16$, and flame extinction occurs at a Reynolds number (*Re*) below 571 for $\varphi = 0.18$ and below 381 for $\varphi = 0.20$. Additionally, a significant amount of unburned hydrogen exits the reactor in all cases. However, with a Pt catalyst coated on the walls, complete H₂ combustion is achieved for $0.10 \le \varphi \le 0.20$, with gas-phase (homogeneous) reactions becoming more prominent at higher Re. In addition, the superadiabatic temperatures are observed close to reactor walls in all studies cases. Moreover, wall radiation and inlet conditions influence combustion kinetics and flame temperature. Under the same conditions, NO_x emissions increase with equivalence ratio but are negligibly affected by the inflow Reynolds number.

Given the high cost and scarcity of noble metal catalysts, this study also focuses on a numerical investigation to determine the best way of coating a platinum catalyst inside a catalytic hydrogen reactor. Various planar and non-planar reactor configurations are examined, and the results show that a reactor combining half and full cylinders is the most

iii

effective in achieving better H_2 conversion. Compared to an equivalent planar catalytic reactor, this non-planar configuration improves H_2 conversion by 30.7%. The findings suggest that enhancing mass and heat convection significantly boosts H_2 conversion. Moreover, the contours of flow parameters and temperatures reveal that cylinders inside the reactor significantly affect the flow near the catalytic surfaces and have benefits in reducing the intensity of super adiabatic temperatures. Additionally, non-planar reactors, with surfaces of improved mass and heat transfer, can achieve up to 50% catalyst savings while maintaining a conversion rate of 2 kg/s per unit of catalytically-coated surface area.

To simulate a realistic catalytic process, Large Eddy Simulations (LES) have been conducted. This represents the first attempt at LES modelling for catalytic monolith reactors to predict catalytic reacting flows. A premixed hydrogen/air mixture at a fuel-lean equivalence ratio of 0.15 and an incoming Reynolds number of 3500 is used for analysis. Both planar and non-planar reactors are studied and compared under the same conditions and with the same platinum-coated surface area. The simulations employed a turbulent kinetic energy sub-grid model and the eddy dissipation concept to model the turbulent catalytic reacting flow. A discrete ordinate model is used to account for radiation heat transfer. The LES results, validated against experimental data, demonstrate that placing cylinders along the reactor enhances convective mass transfer, intensifies catalytic combustion, and enables efficient combustion with less catalytic surface area. Compared to planar models, non-planar reactors exhibited significantly higher H₂ conversion efficiency along the reactor length, allowing for a catalyst savings of nearly 62.5%.

Finally, an experiment was conducted in a catalyst-packed bed tubular reactor to investigate the effect of varying catalyst content (0.3%, 0.5%, or 1.0% Pt in Al₂O₃ pellets and 0.5%, or 5.0% Pd in Al₂O₃ pellets) and catalyst loading (1.0 g, 2.5 g, and 5.0 g). The choice of experimental approach in a packed bed reactor is due to limitations of addressing the effect of catalyst material and amount of catalyst loading in numerical modelling. Measurements were taken in the packed bed reactor across the flow rates ranging from 1 LPM to 5 LPM. The results show that the packed bed with higher Pt/Pd content generates elevated combustion temperatures and demonstrates an effective catalytic performance. Moreover, the pellets with Pt/Pd content, even with a loading of 1 g at low flow rates, exhibit catalytic performance comparable to higher catalyst loadings at different flow conditions.

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Author's declaration

I certify that this thesis, submitted for examination for the PhD degree at the University of Glasgow, is entirely my own work, except where contributions from others are duly acknowledged. It has not been edited by a third party beyond the limits allowed by the University's PGR Code of Practice. This thesis has been produced in accordance with the University of Glasgow's Code of Good Practice in Research.

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Md Nur Alam Mondal January 2025

Contents

Abstract	ii
Acknowledgements	iv
Author's declaration	V
Contents	vi
List of tables	viii
List of figures	ix
Nomenclature	xiv
Publications from this work	xvii
Chapter 1: Introduction	1
1.1 Overview	1
1.1.1 Hydrogen in energy sector	1
1.1.2 Hydrogen combustion systems	5
1.1.3 Catalytic hydrogen combustion (CHC)	11
1.1.4 Intensification of the CHC process	
1.1.5 Research challenges and gaps	14
1.2 Objectives of the research and contributions to knowledge	16
1.3 Thesis structure	17
Chapter 2: Numerical investigation of premixed hydrogen/air combustion	at lean to ultra-
lean conditions and catalytic approach to enhance stability	
2.1 Introduction	
2.2 Computational modelling	
2.2.1 Chemical kinetics	
2.2.2 Validation of the numerical simulations	
2.3 Results and discussion	
2.3.1 Flame stability map in non-catalytic burner	
2.3.2 Catalytic burner combustion	
2.3.2.1 Flame stability	
2.3.2.2 Effect of coupled (hetero-/homogeneous) chemistry	
2.3.2.3 Surface temperatures	

2.3.2.4 H ₂ conversion	39
2.3.2.5 NO _x emission	42
2.4 Conclusions	44
Chapter 3: Enhancing the performance of catalysts in turbulent premixed fuel-lean	
hydrogen/air combustion	46
3.1 Introduction	46
3.2 Modelling of turbulent catalytic reacting flow	49
3.2.1 Governing equations	49
3.2.2 Boundary conditions	52
3.2.3 Solutions algorithm and solver setting	53
3.2.4 Chemical kinetics	55
3.2.5 Mesh resolutions	55
3.3 Validation of the numerical solutions	58
3.4 Results and discussion	61
3.4.1 Planar reactor configurations	61
3.4.2 Conceptual design modifications for optimal catalyst	65
3.4.2.1 Combustion characteristics	65
3.4.2.2 Heat and mass transfer	68
3.4.2.3 H ₂ catalytic conversions	71
3.5 Conclusions	74
Chapter 4: A platinum-coated staggered reactor to intensify lean hydrogen/air combust	ion:
a large eddy simulation study	76
4.1 Introduction	76
4.2 Numerical approach	79
4.2.1 Catalytic reactor configurations	79
4.2.2 Governing equations	82
4.2.3 Solutions algorithm and solver setting	84
4.2.4 Mesh resolutions	85
4.3 Validation of the numerical Solutions	88
4.4 Results and discussion	90
4.5 Discussion	105
4.6 Conclusions	105
Chapter 5: Effect of catalyst quantity on catalytic combustion	107

5.1 Introduction
5.2 Experiments110
5.2.1 Experimental setup and equipment110
5.2.2 Instrumentation
5.2.2.1 Temperature measurement
5.2.2.2 Mass spectrometry (MS)111
5.2.2.3 Data calculation111
5.3 Results and discussion112
5.3.1 Reactor temperatures
5.3.2 H ₂ conversions
5.4 Conclusions
Chapter 6: Conclusions and future work136
6.1 Principal findings136
6.2 Recommendations for the future work
References

List of tables

Table 3.1 Computational conditions	53
Table 3.2 Number of grids tested	56
Table 3.3 Gaseous and catalytic conversion rates at different planar reactor heights	63
Table 3.4 Gaseous and catalytic conversion rates at different catalytic reactor	73
Table 3.5 Pressure drops	74
Chapter 4	
Table 4.1 Simulated cases	80
Chapter 5	
Table 5.1 List of catalyst samples	113
Table 5.2 Thermocouple measurements (°C) for catalysts 0.5% Pt/Al ₂ O ₃	123
Table 5.3 Thermocouple measurements (°C) for catalysts 1% Pt/Al ₂ O ₃	125
Table 5.4 Thermocouple measurements (°C) for catalysts 0.5% Pd/Al ₂ O ₃	127
Table 5.5 Thermocouple measurements (°C) for catalysts 5% Pd/Al ₂ O ₃	129
Table 5.6 Hydrogen conversion (%) measurements for Pt /Al ₂ O ₃ at different catalyst	
loadings	132
Table 5.7 Hydrogen conversion (%) measurements for Pd /Al ₂ O ₃ at different catalyst	
loadings	134

List of figures

Chapter 1

Figure 2.1 Schematic of the catalytic burner geometry (L – burner length, h – gap between
wall)
Figure 2.2 Grid test: Transverse profiles at three selected streamwise locations, (a) hydrogen
mole fraction (X _{H2}) (b) Temperature (T), φ =0.20, T _{in} =312 K24
Figure 2.3 Comparisons of (a) species and temperature profiles at axial positions (b) OH
contour plot (arrow indicates the onset of homogeneous ignition), ϕ =0.28, T _{in} =312 K, Re
=762
Figure 2.4 H_2/air combustion in non-catalytic burner (a) stability map as a function Re (b)
Average value along the burner length (ϕ =0.20, T _{in} =312 K): Left Axis- Mole fraction (X _{avg});
Right Axis- Temperature (T_{avg}). OH mole fraction is taken on surface wall. Re=762 (solid
line); Re=1333 (dashed line)29
Figure 2.5 Contour plots at different Reynolds numbers inside non-catalytic burner for $\boldsymbol{\phi}$
=0.20, T_{in} =312 K (a) hydrogen mole fraction (X _{H2}) (b) Temperature (T)30

Figure 2.6 OH contour plot inside catalytic burner (up to $x/h = 30$) at different Re, T _{in} =312
K
Figure 2.7 Catalytic burner: (a) Left axis: Enthalpy flux at inlet (H), Right axis: Net heat loss
at inlet (Q _{loss} /HoR _a) (b) Average outlet temperature (T _{avg,out}), T _{in} =312 K32
Figure 2.8 Comparison of species mole fraction and temperature distribution on wall
between Coupled (CC) and pure catalytic (PC) chemistry, φ =0.20, T _{in} =312 K (a) Re =190,
(b) Re =762
Figure 2.9 Comparison of species surface coverage on wall between Coupled (CC) and pure
catalytic (PC) chemistry, ϕ =0.20, T _{in} =312 K (a) Re =190, (b) Re =76235
Figure 2.10 Maximum wall temperature using pure catalytic chemistry (PC), T _{in} =312 K
Figure 2.11 Variation of Incident Radiation (Left Axis) and Net Radiation Flux (Right axis),
Coupled chemistry (CC), φ =0.20, T _{in} =312 K at (a) inlet (b) wall
Figure 2.12 (a) H_2 conversion at different Re, (b) H_2 and H_2O distribution at $x/h = 0.5$,
Coupled chemistry (CC), φ =0.20, T _{in} =312 K
Figure 2.13 Residence Time as a function of Re using coupled chemistry (CC), φ =0.20,
T _{in} =312 K40
Figure 2.14 Local Sherwood Number (Sh) along the burner length for H ₂ , Coupled chemistry
(CC), φ =0.20, T _{in} =312 K42
Figure 2.15 Emission as a function of φ using coupled chemistry (CC) (a) NO _x (b) Thermal
NO, $Re = 1333$, $T_{in} = 600$ K43

Figure 3.1 Schematics of the various catalytic reactors. The red colour denotes the catalytic
surface in the catalytic zone
Figure 3.2 Effect of grid resolution for the configurations of C2, C3 and C4, (a)
temperature on catalytic surface, $T_{cat}(c, e)$ midplane temperature inside the reactor, $T(b, d, cat)$
f) transverse profile of H ₂ mass fraction at location $x=7.5$ mm and 9 mm
Figure 3.3 Comparisons of species and temperature mean profiles at different axial
positions at φ =0.18, T_{in} =300 K, Re =15390
Figure 3.4 OH contour (ppmv) (arrow indicates the onset of gas-phase ignition) at φ =0.18,
T_{in} =300 K and Re =1539060

Figure 3.5 Planar catalytic reactor: (a) left axis: H ₂ Conversion; right axis: temperature
distribution on catalytic wall (T _w) (b) H ₂ and H ₂ O distribution at x=10 mm, φ =0.15,
T_{in} =300 K, Re =420062
Figure 3.6 Surface coverages (a) H(s) (b) O(s) (c) OH(s) , ϕ =0.15,64
Figure 3.7 Contours of (a) Y_{H2} (b) Y_{H2O} (c) Y_{OH} and (d) T for reactors C1, C2, C3 and
C4. Reactor length from x=0 to 57.5-mm is presented, φ =0.15, T_{in} =300 K66
Figure 3.8 Streamlines for reactors C2, C3 and C4. Reactor length from $x=2.5$ -mm to 25.5-
mm is presented67
Figure 3.9 Nusselt Number (Nu) on catalytic surfaces for configurations C1, C2, C3 and
C469
Figure 3.10 Sherwood Number (<i>Sh</i>) of H_2 on catalytic surfaces for configurations C1, C2,
C3 and C470
C3 and C4
C3 and C4

Figure 4.1 Schematics of the (a) planar and (b) non-planar reactors79
Figure 4.2 Example of grid generation
Figure 4.3 Effect of grid resolution for the catalytic planar configuration (Case 3). Time
averaged transverse profiles of (a) U_m and k, (b) T_m and T_{rms} , and (c) Y_{H2} and $Y_{H2,rms}$ at x/H
=7.5, T_{in} =300 K, φ =0.15 and Re =350086
Figure 4.4 Effect of grid resolution for the catalytic non-planar configuration (Case 4). Time
averaged transverse profiles of (a) U_m and k , (b) T_m and T_{rms} , and (c) Y_{H2} and $Y_{H2,rms}$, at
$x/H=7.23$, $T_{in}=300$ K, $\varphi=0.15$ and $Re=3500$
Figure 4.5 Case C1 (reacting): LES comparison at various streamwise locations for the time-
averaged mean profiles of (a) U_m and k , (b) X_{H2} and X_{H2O} , and (c) T_m , at $T_{in}=300$ K, $\varphi=0.18$,
<i>Re</i> =15390
Figure 4.6 Case C2 (non-reacting): LES comparison for the time-averaged mean velocity
(U_m and V_m) and Reynold stresses (R_{UV}) at $x/H=6.51$, $Re=21304$ 90
Figure 4.7 Case 3 (reacting): Contours of (a) instantaneous x-velocity, U, (b) time-
averaged mean x-velocity, U_m and (c) its fluctuations, U_{rms} onto the xy-plane at $z/H =$
0.75. Transverse profiles of time-averaged (d) U_m (e) k (f) U_{rms} (h) V_{rms} (h) W_{rms} and (i)
R_{UV} (or $U_{rms}V_{rms}$) at five streamwise locations

Figure 4.8 Case 3 (non-reacting): Transverse profiles of time-averaged (a) U_m (b) k (c)
U_{rms} (d) V_{rms} (e) W_{rms} and (f) R_{UV} (or $U_{rms}V_{rms}$) at five streamwise locations. The number
from 1 to 5 indicates axial locations shown in Figure 4.7(a)92
Figure 4.9 Case 3 (reacting): Instantaneous contour of (a) T (b) Y_{H2} (c) Y_{H2O} (d) Y_{OH} onto
the xy-plane at $z/H = 0.75$; Transverse profile of (e) T_m (f) T_{rms} (g) $Y_{H2,m}$ (h) $Y_{H2,rms}$, the
number from 1 to 5 indicates axial locations shown in Figure 4.7(a)95
Figure 4.10 Case 3 (reacting): (a) The predicted locally averaged (upper and lower
surfaces) catalytic conversions (C) and the integrated (over the reactor flow height) gaseous
conversions (G) along the streamwise direction. (b) Local distribution of Nu , Sh , τ_w and T_w
on reactor inner surface
Figure 4.11 Case 4 (reacting): Contours of (a) instantaneous x-velocity, U (b) time-
averaged mean x-velocity, U_m and (c) its fluctuations, U_{rms} onto the xy-plane at $z/H = 0.75$
Transverse profiles of time-averaged (d) U_m (e) k (f) U_{rms} (g) V_{rms} (h) W_{rms} and (i) R_{UV}
(or $U_{rms}V_{rms}$) at six streamwise locations
Figure 4.12 Case 4 (reacting): Instantaneous contour of (a) T (b) Y_{H2} (c) Y_{H2O} (d) Y_{OH} onto
the xy-plane at $z/H = 0.75$; Transverse profile of (e) T_m (f) T_{rms} (g) $Y_{H2,m}$ (h) $Y_{H2,rms}$, the
number from 1 to 6 indicates axial locations shown in Figure 4.11(a) (i) The predicted
locally averaged (upper and lower cylindrical surfaces) catalytic conversions (C) and the
integrated (over the reactor flow height) gaseous conversions (G) along the streamwise
direction101
Figure 4.13 Case 4 (reacting): local distribution of (a) Nu , (b) Sh and (c) τ_w on catalytic
cylindrical surfaces
Figure 4.14 Hydrogen conversion (%) along the length of the reactor104

Figure 5.1 (a) monolithic catalytic reactor (b) catalyst packed bed reactor. Figures from	n
[184,185] are reused with permission.	109
Figure 5.2 Schematic of experimental setup	110
Figure 5.3 Reactor wall temperatures (°C) obtained from IR thermal camera, 0.5%	
Pt/Al ₂ O ₃	115
Figure 5.4 Reactor wall temperatures (°C) obtained from IR thermal camera, 1.0%	
Pt/Al ₂ O ₃	116
Figure 5.5 Reactor wall temperatures (°C) obtained from IR thermal camera, 0.5%	
Pd/Al ₂ O ₃	117

Figure 5.6 Reactor wall temperatures (°C) obtained from IR thermal camera, 5.0%
Pd/Al ₂ O ₃
Figure 5.7 Catalyst (0.5% Pt/Al ₂ O ₃) bed temperatures at different locations for loading: (a)
1g (b) 2.5g (c) 5g
Figure 5.8 Catalyst (1% Pt/Al ₂ O ₃) bed temperatures at different locations for loading: (a)
1g (b) 2.5g (c) 5g
Figure 5.9 Catalyst (0.5% Pd/Al ₂ O ₃) bed temperatures at different locations for loading: (a)
1g (b) 2.5g (c) 5g
Figure 5.10 Catalyst (5% Pd/Al ₂ O ₃) bed temperatures at different locations for loading: (a)
1g (b) 2.5g
Figure 5.11 Effect of catalyst loading on hydrogen conversion for (a) 0.5% Pt/Al ₂ O ₃ (b)
1% Pt/Al ₂ O ₃
Figure 5.12 Effect of catalyst loading on hydrogen conversion for (a) 0.5% Pd/Al ₂ O ₃ (b)
5% Pd/Al ₂ O ₃

Nomenclature

d	Cylinder diameter (m)
D_m	Mass diffusivity (m ² /s)
Ε	Total energy (W)
G	Radiation intensity (W/m ²)
h_c	Heat transfer coefficient (W/m ² K)
HoR	Heat of reaction (W)
J	Diffusion flux of species (kg/m ² s)
k_c	Mass transfer coefficient (-)
L	Length (m)
Le	Lewis number (-)
'n	Mass flowrate (kg/s)
М	Molecular weight (kg/kmol)
N_s	Total of number of surface species (-)
N_g	Total number of gas species (-)
Nu	Nusselt number (-)
р	Pressure (N/m ²)
Р	Cylinder pitch (m)
Pr	Prandtl number
q	Heat flux (W/m ²)
Q_{loss}	Net heat loss (W)
Q_{rad}	Radiation heat (W)
q_{rad}	Radiation heat flux (W/m ²)
Re	Reynolds number (-)
Ś	Molar production rate (mol/m ² s)
S	Surface area (m ²)
Sc	Schmidt number
Sh	Sherwood number (-)
Т	Temperature (K)
u_t	Friction velocity (m/s)
ŵ	Reaction rate of gas species (kg/m ³ s)
x	Streamwise coordinate (m)

X	Mole fraction (-)
Y	Mass fraction (-)
у	Transverse coordinate (m)
Ζ	Spanwise coordinate (m)

Greek symbols

ρ	Density (kg/m ³)
μ	Dynamic viscosity (kg/ms)
arphi	Equivalence ratio (-)
λ	Thermal conductivity (W/mK)
Г	Surface site density (mol/cm ²)
θ	Surface coverage (-)

Subscripts

a	Actual
adb	Adiabatic
avg	Average
cat	Catalyst
G	Fluid
in	Inlet
m	Mean
max	Maximum
out	Outlet
rms	Root mean square
S	Solid
W	Wall surface

Superscripts

Filtered variable

-

Abbreviations/ Acronyms

CHC	Catalytic Hydrogen Combustion
CC	Couple Chemistry
СОР	Conference of the Parties
CCS	Carbon Capture and Storage
DO	Discrete Ordinates
DNS	Direct Numerical Simulations
EDC	Eddy Dissipation Concept
FID	Final Investment Decision
FVM	Finite Volume Method
ICE	Internal Combustion Engines
LES	Large Eddy Simulation
MID	Multiple Ion Detection
NZE	Net Zero Emissions
PC	Pure Catalytic Chemistry
QMS	Quadrupole Mass Spectrometer
RANS	Reynolds-Averaged Navier Stokes
S/V	Surface to Volume Ratio
SGS	Sub Grid Scale

Publications from this work

Journal articles

Mondal, M. N. A., Karimi, N., Jackson, S.D., Paul, M.C., (2024) *A platinum-coated staggered reactor to intensify lean hydrogen/air combustion: a large eddy simulation study*, Fuel, Volume 381, Part B, 2025, 133386, ISSN 0016-2361, <u>https://doi.org/10.1016/j.fuel.2024.133386</u>

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Research poster

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Chapter 1: Introduction

1.1 Overview

Global energy policy is now firmly focused on transitioning away from fossil fuels and reducing or eliminating net greenhouse gas emissions in the energy sector. Currently, most of the world's energy used in transportation, power generation, and heating is produced through combustion. Consequently, advancing combustion research is essential to achieving a carbon-free, low-emission, and sustainable energy sector. In this regard, hydrogen combustion is widely recognised as a promising and environmentally friendly energy solution. This introductory section provides an overview of the current status and future prospects of hydrogen in the energy sector, along with a discussion of hydrogen combustion systems and advanced techniques for achieving low or ultra-low NO_x emissions in hydrogen combustion.

1.1.1 Hydrogen in energy sector

The utilisation of hydrogen as a cleaner fuel is accelerating across the global energy sector, driven by efforts to replace fossil fuels and combat climate change [1,2]. The significance of this transition is highlighted in the International Renewable Energy Agency (IRENA) report as follows:

"In this 1.5°C scenario, the global hydrogen production would need to expand by almost five times, to 614 megatonnes of hydrogen per year, to reach 12% of final energy demand by 2050, also shifting from a major source of greenhouse gas emissions to a low-emission energy carrier. Green hydrogen, produced from renewables, is expected to represent the bulk of the production" [3]

Consequently, many countries have articulated their strategies for decarbonising the energy sector, with hydrogen playing a central role in most of these plans. For example, the UK government released its hydrogen strategy [4], which outlines a groundbreaking vision to establish a leading hydrogen economy and a vibrant energy sector. This initiative aims to reach a hydrogen production capacity of 5 gigawatts, contribute £900 million to the UK economy, support over 9,000 jobs, and unlock £4 billion in investment in the UK by 2030.



Figure 1.1 The role of hydrogen in connecting various energy sectors reused with permission from elsevier [5]

With an energy density of 120 MJ/kg, nearly three times that of hydrocarbons, hydrogen presents a strong potential as a fuel source [6]. Hydrogen, which can be produced from both fossil fuels and renewable sources [7] as shown in Figure 1.1, plays a critical role in linking hydrogen-consuming sectors such as ammonia production and synthetic fuels with key industries including transportation, power generation, gas grids, residential heating, and energy storage [5,8]. Hydrogen production currently amounts to around 95 million tonnes (Mt) globally, with the majority being grey hydrogen, as fossil fuels account for approximately 99.3% of total hydrogen production, as shown in Figure 1.2. Furthermore, this scenario is expected to change in the coming years due to commitments made at the latest Conference of the Parties (COP28) in Dubai, aimed at transitioning energy systems away from fossil fuels to meet the goals of the Paris Climate Agreement. As a result, many countries have set ambitious hydrogen production targets as part of their net-zero 2050 strategies. According to a recent report from the Hydrogen Council [9], the global supply of clean hydrogen is expected to come online at a rate of 2-3 Mt per year by 2024, with projections of increasing to 6-8 Mt per year by 2027 and 12-18 Mt per year by 2030. For this, the electrolysis and fossil fuels with CCS technology, as illustrated in Figure 1.2, have been identified in the literature [10-13] as a promising method for clean hydrogen production. Furthermore, to achieve net-zero targets, the number of globally announced hydrogen projects has increased to 1572, with 1125 projects expected to begin commercial operation by 2030 [9]. The planned production capacity of 48 Mt per year in these projects

has been announced through 2030, with 75% focused on clean hydrogen production. The total investment committed to these projects amounts to 680 billion USD through 2030. However, hydrogen production costs remain a challenge [14]. As shown in Figure 1.3, the levelised cost of hydrogen production was at its lowest in 2021 but increased in 2022 due to the impacts of the COVID-19 pandemic [15]. In 2021, the levelised costs of hydrogen ranged from 1.0-3.0 USD/kg for hydrogen produced from unabated fossil fuels and 1.5-3.6 USD/kg for hydrogen produced from fossil fuels with carbon capture and storage (CCS).



Figure 1.2 Hydrogen production by sector (a) 2022 (b) 2030



Figure 1.3 Levelised cost of per kg hydrogen production [15]

Under the Net Zero Emissions (NZE) scenario, with technological advancements and largescale deployment, production costs are expected to decline, falling to 0.5-2.7 USD/kg for hydrogen from unabated fossil fuels and 2.0-8.0 USD/kg for hydrogen from fossil fuels with CCS and renewable sources, respectively.

Figure 1.4 highlights the current landscape of hydrogen use across various sectors. While global hydrogen consumption continues to grow, it remains concentrated in traditional applications such as industry and refining. The uptake of hydrogen in critical sectors like transport, power generation, and synthetic fuels production—key drivers of the energy transition—remains minimal, accounting for just 0.1% of global energy demand. According to the reports [15,16], hydrogen use is projected to increase by 6% annually, reaching over 150 Mt by 2030 and 430 Mt by 2050 as shown in Figure 1.4, with the majority of growth coming from transport, power, synthetic fuels, and buildings. To achieve the NZE targets, it is crucial to accelerate the adoption of hydrogen-fuelled technologies across all sectors. In transport, for instance, hydrogen use grew by 45% in 2022 and is expected to reach 8 Mt by 2030, with 50% allocated for road transport and the remaining portion for shipping [15]. Additionally, around 8 Mt of hydrogen will be used in the production of ammonia and synthetic fuels for aviation and maritime applications.



Figure 1.4 Hydrogen consumption by sector [16]

However, hydrogen's role in power generation remains limited, contributing less than 0.2% to the global energy mix [15], where it is typically used in conjunction with other gases. On the positive side, technologies such as fuel cells [17,18], internal combustion engines (ICE) [19], and gas turbines [20,21] are being modified to run on hydrogen-rich fuels or even pure hydrogen, enabling commercial deployment. In the building sector, low-carbon alternatives

are being utilised to meet climate goals, such as renewable electricity for heat pumps [22], district heating from renewable or waste sources [23], and rooftop solar or local wind turbines [24] for electricity generation. For hydrogen-fuelled technologies [25] to compete, further research and innovation are required. Hydrogen use in buildings is projected to surpass 1.0 Mt by 2030, but this is still negligible in the NZE scenario, contributing only 0.14% to the total energy demand [15].

1.1.2 Hydrogen combustion systems

A key advantage of hydrogen is its potential as a highly promising fuel, driven by extensive innovation and research aimed at adapting hydrogen for use in combustion systems. It provides heat and power while offering a viable pathway to decarbonise a wide range of industrial, residential, and automotive applications through combustion. During hydrogen combustion in air, hydrogen reacts with oxygen, as outlined in belowbelow, producing water vapor and releasing energy.

$$H_2 + \frac{1}{2}(O_2 + 3.76N_2) \to H_2O + 1.88N_2; \ \Delta H_{T_{\infty,H_2O}}^{f,gas} = -241\frac{KJ}{kg}$$
(1.1)

However, hydrogen combustion in air is not entirely emission-free. At high temperatures, NO_x (nitrogen oxides) is formed due to the reaction between nitrogen and oxygen molecules in the combustion flue gas. NO_x refers to a combination of nitrogen oxides and NO (nitric oxide) and NO_2 (nitrogen dioxide) are the main contributors.

NO formation in hydrogen/air combustion occurs through three primary mechanisms: thermal NO (Zeldovich mechanism), N₂O and NNH routes. The thermal NO mechanisms [26] mentioned below is significant at high combustion temperatures. The initial step involves the reaction between oxygen atom and nitrogen gas. This reaction has an activation energy of 75 kcal/mol and produces NO and a nitrogen atom (N). Subsequently, the nitrogen atom reacts with hydroxyl radicals (OH) and O₂ to form additional NO.

$$0 + N_2 \leftrightarrow NO + N \tag{1.2}$$

$$N + O_2 \leftrightarrow NO + O \tag{1.3}$$

$$N + OH \leftrightarrow NO + H \tag{1.4}$$

The N₂O and NNH pathways for NO formation are important at low combustion temperatures or high pressures in fuel-lean operating conditions. For the details of reaction mechanisms, refer to the work of Glarborg *et al* [26]. However, the key reaction steps for these two pathways are:

N₂O pathway:

$$N_2 O + M \leftrightarrow N_2 + O + M \tag{1.5}$$

$$N_2 O + H \leftrightarrow NO + NH \tag{1.6}$$

$$N_2 0 + 0 \leftrightarrow 2N0 \tag{1.7}$$

$$N_2 O + H \leftrightarrow N_2 + OH \tag{1.8}$$

$$N_2 0 + 0 \leftrightarrow N_2 + O_2 \tag{1.9}$$

NNH pathway:

$$NNH \leftrightarrow N_2 + H$$
 (1.10)

$$NNH + 0 \leftrightarrow N_2 0 + H \tag{1.11}$$

$$NNH + 0 \leftrightarrow N_2 + 0H \tag{1.12}$$

$$NNH + 0 \leftrightarrow NH + NO$$
 (1.13)

$$NNH + O_2 \leftrightarrow N_2 + HO_2 \tag{1.14}$$

 NO_2 formation occurs with the presence of hydroperoxyl radical (HO₂) which is an important intermediate in hydrogen combustion [27]. The HO₂ react with NO to form NO₂ in the following reaction:

$$NO + HO_2 \leftrightarrow NO_2 + OH$$
 (1.15)

However, NO_x is one of the major air pollutants and has been found to be responsible for asthma and several respiratory issues, significantly impacting overall human health [28]. Moreover, NO_x is a major contributor to smog formation, acid rain, and ozone layer depletion [28]. Therefore, the reduction of NO_x emissions is one of the major aspects of hydrogen combustion systems. Hydrogen combustion systems can be categorised based on their applications, as depicted in Figure 1.5. These include:

a) Hydrogen Internal Combustion Engine (H₂ ICE): These engines burn hydrogen in a manner similar to traditional spark ignition engines. Some challenges faced by these engines reported in literature [29,30] include combustion instabilities such as pre-ignition, improper ignition timing leading to knocking or misfiring, and the generation of NO_x emissions at high temperatures. Extensive research [28,29] is being conducted on H₂ ICE design parameters to ensure safe vehicle operation while maximising engine power and controlling NO_x emissions.

- b) Hydrogen gas turbine: Hydrogen gas turbines offer a promising and versatile solution for the power, aviation, and industrial applications to meet the world's future energy demands [33–35]. However, transitioning to hydrogen gas turbines requires significant modifications to existing infrastructure, refuelling systems, and storage facilities [36]. A significant challenge of utilising hydrogen in gas turbines is hydrogen embrittlement causing sudden blade failure[37,38]. This issue arises when hydrogen penetrates the blade material, resulting in increased embrittlement that leads to cracking. Additional challenges include high flame speeds leading to flashbacks, as well as hydrogen storage and leakage concerns that raise safety issues [39]. Furthermore, hydrogen combustion in turbines can reach adiabatic temperatures of around 2500°C, resulting in NO_x emissions, turbine material degradation, and reduced overall efficiency [39]. On the positive side, ongoing research and innovation are focused on making gas turbine operation more sustainable, safe, and efficient [34,36].
- c) Hydrogen burner: Hydrogen burners hold significant potential to replace fossil fuels for heating in buildings, industrial processes, and cooking applications [25,40]. However, challenges such as the risk of flashback and elevated NO_x emissions pose major obstacles to their practical and safe operation [36,37]. Studies have shown that these burners generate substantially higher NO_x emissions compared to natural gas burners, primarily due to the higher combustion temperatures [43].



Figure 1.5 Hydrogen combustion systems [31,39,44]



Figure 1.6 NO_x emission characteristics, combustion temperature, and efficiency as functions of the equivalence ratio: illustrative performance derived from an internal combustion engine [45,46]. Reuse from [46] with permission from elsevier.

As discussed above NO_x emissions are crucial in most hydrogen combustion systems. If NO_x emissions are not effectively controlled, it could hinder the widespread adoption of hydrogen in combustion systems, especially as the global community strives to achieve net-zero emissions under the Paris Climate Agreement. As a result, advancements in combustion techniques are essential to bring NO_x emissions within international regulatory limits.

Generally, NO_x emission in hydrogen combustion is influenced by the hydrogen/air mixing ratio and combustion temperature. The relationship among equivalence ratio (the ratio of the actual fuel-to-air ratio to the stoichiometric fuel-to-air ratio) and combustion temperature and NO_x emission is illustrated in Figure 1.6. Although this relationship is derived from hydrogen internal combustion engines, it is applicable for understanding NO_x emission characteristics in other hydrogen combustion systems. As shown, NO_x values are highest near stoichiometric conditions, where combustion temperatures reach their peak.

In this work, the focus is on developing low NO_x in hydrogen burners for heating applications. Several methods are suggested in the literature [47–50] for low NO_x hydrogen burners. These are as follows:

- reducing combustion temperature
- using fuel-lean mixtures
- reducing the excess O₂ in fuel/air mixture
- increasing the mixing/ turbulence to reduce the residence time of flue gas
- adding diluents like N₂, He or H₂O



(c) Catalytic porous burner (non-premixed) [50]





c) Primary catalytic diffuser combustion disk SiC foam 80 ppi, Pt coated



e) Air and H 2 diffuser disk SiC foam 40 ppi



b) H $_2$ diffuser disk, for even distribution of H $_2$ SiC foam 100 ppi



d) Air supply tubes and horizontal distribution



f) Secondary catalytic diffuser combustion disk SiC foam 60 ppi, Pt coated



(d) Catalytic honeycomb burner (premixed) [53]

Figure 1.7 Low NO_x burner: (a) Dry low NO_x (DLN) burner (b) Porous inert medium (PIM) burner (c) Catalytic porous burner (non-premixed) (d) Catalytic honeycomb burner (premixed). Burner configurations from [50-53] are reused with permission.

However, the development of low NO_x burners is a major area of research for various fuels and has been carried out for many years. Some of the recent technologies of low NO_x burner for hydrogen fuel are summarised below.

DLN (dry low NO_x) burner: DLN burner as shown in Figure 1.7 (a) is based on lean premixed combustion of reducing NO_x and combustion temperature, and uses swirling mixing technologies. Key features of DLN burners are the primary (premixed) and secondary (non-premixed) fuel mixing. However, there have been reports of operational issues, such as lean blow-off, flashback, and increased combustion instabilities when operating at leaner combustion conditions.

PIM (porous inert medium) burner: The PIM burner in Figure 1.7 (b) utilises a porous block that facilitates a recirculation heat transport phenomenon. This design helps to lower combustion temperatures and reduce NO_x emissions. Additionally, these burners offer improved flame stabilisation, higher power throughput, and a wider flammability range [54–57]. However, there are several practical design challenges associated with PIM burners, including the selection of suitable porous materials, determining the appropriate length of the porous bed, incorporating external heat exchangers, and ensuring functionality at elevated pressures.

Catalytic burner: Catalytic burners as illustrated in Figure 1.7 (c) and (d) utilises catalyst material within its design to control the combustion process. These burners operate both on premixed and diffusion principles as shown in the Figure 1.7 (c) and (d), respectively. Additionally, catalytic burners demonstrate great effectiveness in reducing NO_x emissions and have been extensively researched across various applications. Among the applications, catalyst use in honeycomb, bluff body and porous burners is common [58–60]. However, due to the ability of catalytic burners to oxidise hydrogen completely at low temperatures, catalytic burners are gaining popularity. A detailed discussion of the catalytic burner technique for hydrogen combustion follows in the next section.

1.1.3 Catalytic hydrogen combustion (CHC)

Catalytic combustion, first introduced by Pfefferle *et al.* [61,62] in 1970, involves the use of a catalyst to accelerate the combustion reaction without altering the chemical equilibrium. Since its inception, catalytic combustion has been widely adopted as an efficient method for burning lean mixtures of various hydrocarbon fuels, resulting in lower emissions. [63–69]. In particular, catalytic hydrogen combustion (CHC) produces zero carbon emissions due to the absence of carbon in the fuel. CHC systems as shown in Figure 1.5 can be classified into two types: hybrid (500-1500°C) [70–77] and catalytic combustion (< 500°C), based on the combustion mode. In hybrid systems, both homogeneous and heterogeneous reactions occur, whereas, in catalytic combustion, the process is entirely heterogeneous, as the reaction temperature remains below the autoignition temperature of 583°C [44].

Catalytic combustion is a well-established and promising technique for heat and power generation, used in various applications [34,39,75]. This technique provides complete combustion under diverse conditions while significantly reducing emissions—something

that is difficult to achieve with conventional combustion, even with design modifications [78,79]. In catalytic, the catalyst facilitates the hydrogen oxidation reaction without being consumed. The fundamental principles of catalytic combustion and its behaviour across different reactor systems have been extensively reviewed in the literature [44,80–82]. Over the years, both experimental and numerical studies have advanced the understanding of catalytic processes [53,68,83–85]. Currently, innovations and advancements in catalytic combustion are rapidly evolving, particularly in integrating this technology into combustion systems such as gas turbines, furnaces, and boilers [21,34,86].

Catalytic combustion implemented in combustion systems, as illustrated in Figure 1.8, can be classified into two types: fuel-lean catalytic combustion and fuel-rich catalytic combustion [87]. Fuel-lean catalytic combustion can further be divided into two subtypes: partial catalytic conversion, where part of the fuel undergoes catalytic conversion and the remainder is combusted in the gas phase, and total catalytic conversion, which is a purely catalytic process [88]. In fuel-rich catalytic combustion, the catalytic conversion occurs under fuel-rich conditions, and the unburned fuel is subsequently combusted in the gas phase with the help of bypass air. The selection of the appropriate catalytic combustion technique depends on the specific application and operating conditions.

However, total catalytic conversion under fuel-lean conditions is effective for hydrogen/air combustion at low temperatures and significantly reducing NO_x . In this case, the catalytic unit plays a critical role, as most catalysts used are noble metals, which are expensive. Additionally, the catalytic performance is also essential, as it varies based on active metal, burner design and catalyst loading. Among the various burner designs, monolithic, porous and packed bed burners are the most commonly used in catalytic combustion systems, as reported in the literature [58,68,89,90]. In a monolithic structure, the catalyst is applied to the surfaces of parallel channels, while in a porous medium, the catalyst is coated onto the surfaces of the pores. In a packed bed burner, active sites are applied to the surfaces of support pellets. Despite the differences in physical design, these burners operate on similar principles, facilitating catalytic reactions by maximising the contact area between the reactants and the catalyst. However, in this work, particular emphasis is placed on CHC in monolithic burners and their efficacy in reducing NO_x under fuel-lean conditions.

However, due to the strong temperature dependence of NO_x formation, low-temperature CHC burners have gained significant attention for domestic and commercial heating systems [82,91,92]. Among the CHC burners, the honeycomb monolith structure [93–96] is simple

in design, effective in fuel conversion and reducing NO_x . Therefore, in this work, particular emphasis is placed on CHC in monolithic burners and their efficacy in reducing NO_x under fuel-lean conditions.



Figure 1.8 Catalytic combustion systems in power generation (a) Fuel-lean catalytic conversion: (a1) Partial catalytic conversion (a2) Total catalytic conversion (b) Fuel-rich catalytic conversion

1.1.4 Intensification of the CHC process

The key components of a monolith burner are the fuel/air mixing technique, the catalyst material, catalyst loading, and the burner design. The catalyst plays a crucial role in fuel consumption and overall combustion efficiency. However, the major limitation of this burner lies in its reliance on costly catalyst materials. While noble metals like platinum (Pt), palladium (Pd), and rhodium (Rh) deliver excellent combustion performance, their high cost

and limited availability hinder the widespread use of catalytic burners. Recent advances [82,92,97,98] in CHC research focus on two main areas: developing more cost-effective catalyst materials and optimising reactor designs to improve mass and heat transfer, thereby enhancing catalytic efficiency. The former requires significant exploration to discover materials that offer both low cost and high catalytic activity. Encouragingly, numerous studies [44,99,100] are underway to discover alternative catalysts suitable for catalytic combustion. Despite the challenges posed by the reliance on noble metals, current research efforts are geared toward minimising the use of these expensive materials while maintaining optimal catalytic performance.

The catalytic combustion reaction is typically limited by mass transport, meaning the reaction rate is much faster than the transport of reactants to the catalytic surface. In this context, the intensification of the catalytic combustion process involves strategies to enhance mass and heat transport towards the catalytic walls and increase the reactant-catalyst interaction, thereby improving the performance of the catalytic reaction. Importantly, the main focus of catalytic intensification in this work is to minimise the use of noble catalysts in the catalytic process and reduces the overall costs of the catalytic reactor.

1.1.5 Research challenges and gaps

The monolithic catalytic reactors reported [66,69,101,102] in the literature are planar in design. Recent studies on planar burners have primarily focused on the effects of flow rates, equivalence ratio, preheat, pressure, and wall temperatures on the CHC process [67,68,76,103,104]. The CHC process occurs on the catalyst-coated surface of the burners and becomes more complex when gas-phase combustion is involved under varying operating conditions. To intensify the catalytic process in these burners, modifying the burner design, particularly the catalytic surface, is essential. Some studies [105–107] have explored design changes, such as incorporating cavities and waviness on the catalytic surface, showing significant improvements in the catalytic process. However, the fundamental understanding of enhancing the catalytic process in design modifications are not fully understood. Additionally, the behaviour of the catalytic process in these modified designs under extremely fuel-lean conditions, beyond flammability, remains unclear. Therefore, a comprehensive investigation into the underlying physics of the CHC process across various design conditions in monolithic burners is needed. Additionally, studies on NO_x emissions in these design conditions are limited, and essential for implementing CHC burners in diverse applications.

The most effective way to investigate the CHC process in a monolithic burner is through experimental methods. However, due to the small dimensions of a monolithic burner, accurately measuring data close to the catalytic surface is quite challenging. As a result, most research [58,108–112] on monolithic burners relies on numerical approaches to predict the CHC process. For analysing the CHC process in a laminar flow regime, the numerical model is well-developed and widely used. However, in practical scenarios, catalytic burners are mostly operated at higher Reynolds numbers to achieve maximum throughput. Surprisingly, there have been few attempts [102,113–115] to investigate the CHC process in a turbulent flow regime. Because the complexity of CHC processes increases significantly when combustion involves turbulence at high flow velocities. Therefore, the numerical modelling in this case becomes challenging to capture the flow turbulence and predict the realistic CHC process. Nevertheless, advanced numerical modelling techniques like RANS and LES are essential for predicting the catalytic process effectively in real-world applications. Appel et al. [102,116] used the RANS model to simulate the CHC process and demonstrate its capabilities to predict their experimental. To the best of the author's knowledge, there have been no reports of LES modelling detailing the CHC process. However, some efforts [113-115] have been made using DNS modelling to study the CHC process in the planar monolithic reactor.

Moreover, minimising the use of expensive catalyst materials is essential for the broader adoption of CHC burners, and this aspect has not been extensively explored in the literature. Enhancing the interaction of reactants with the catalytic surface is critical and requires thorough investigation to reduce catalyst consumption within the burner. However, given the limitations and high costs associated with noble metal catalysts, fundamental research in the above-mentioned unexplored area is needed to optimise CHC burner designs with minimal reliance on these expensive materials. The present work focuses on investigating the catalytic combustion in monolith burners using premixed lean to ultra-lean hydrogen/air mixtures. This mixture is selected due to its ability to generate useful combustion temperatures for heating applications while being essential for achieving zero NO_x emission systems. The work first numerically explores the underlying physics of the catalytic process with these mixtures, addressing a gap in the literature regarding NO_x emissions. Additionally, this work examines the intensification of the catalytic process through variations in monolith burner design, an area also lacking in existing research. Notably, the reactor design proposed in this study has not been explored before. However, the numerical modelling of the catalytic process is both critical and challenging, particularly when turbulent flow is introduced in catalytic reactions due to burner design. Therefore, further research using expensive numerical techniques is vital for accurately predicting real-world catalytic processes. Furthermore, studying catalytic combustion with minimal catalyst usage is crucial for ensuring cost-effectiveness and broadening its applicability. This aspect is not extensively explored in the literature and necessitates experimental investigation to understand the impact of catalyst quantity on the CHC process. In light of these considerations, the present work aims to achieve the following novel objectives:

- To numerically investigate the catalytic combustion of premixed lean to ultra-lean hydrogen/air mixtures within a platinum-coated planar burner, achieving near-zero NOx emissions.
- To numerically analyse the impact of reactor design on the intensification of catalytic combustion in both planar and non-planar burners.
- To predict the catalytic intensification processes in turbulent reacting flows for both planar and non-planar burners using Large Eddy Simulation (LES).
- To experimentally assess the performance of catalytic combustion in a catalyst-packed bed burner by varying catalyst loading.

1.3 Thesis structure

Chapter 1: This study on catalytic hydrogen combustion begins with an introductory chapter that highlights the necessity of hydrogen as a replacement for fossil fuels, along with a discussion of the global hydrogen production landscape and its growing demand in the energy sector. The primary objective of the introduction is to emphasise the importance of adopting hydrogen in combustion systems and to explore the potential of catalytic combustion technology as a key solution for replacing fossil fuels, particularly in domestic and commercial heating applications.

Chapter 2: Numerical simulations of both catalytic and non-catalytic hydrogen combustion in a planar burner are performed by varying fuel concentration to ultra-lean conditions. The simulations are further extended to investigate the transition of combustion modes (from catalytic to coupled reactions) at different hydrogen/air mixtures. Longer catalytic burners are analysed to achieve complete hydrogen combustion at varying inflow Reynolds numbers. NO_x emissions are evaluated.

Chapter 3: Further simulations explore the intensification of the catalytic process in both planar and non-planar catalytic burners, incorporating practical design modifications. The calculated Nusselt and Sherwood numbers highlight the effectiveness of the catalytic process across different burner configurations. Conversion rates are also quantified to compare the performance of the various catalytic burners analysed in this study.

Chapter 4: Large Eddy Simulation (LES) is employed to predict the complex catalytic processes in turbulent reacting flows within both planar and non-planar catalytic burners. Experimental data is used to validate the LES results. Key turbulent parameters, including shear stress, Nusselt number, and Sherwood number, are assessed to characterise the turbulent behaviour in the catalytic combustion process.

Chapter 5: Experiments on catalytic combustion in a tubular catalyst-packed bed burner using alumina-supported pellets are conducted to evaluate the effectiveness of catalytic performance by varying catalyst loading. The experimental results highlight the impact of catalyst loading and catalyst temperatures on the catalytic combustion process.

Chapter 6: The study's overall results are summarised, and potential directions for future research are suggested.
Chapter 2: Numerical investigation of premixed hydrogen/air combustion at lean to ultra-lean conditions and catalytic approach to enhance stability

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Abstract

Premixed combustion of hydrogen/air over a platinum (Pt) catalyst is numerically investigated in a planar channel burner with the aim of stabilising the flame at lean to ultralean conditions. A steady laminar species transport model is examined in conjunction with elementary heterogeneous and homogeneous chemical reaction schemes and validated against experimental results. A stability map is obtained in a non-catalytic burner for the equivalence ratios (φ) of 0.15-0.20, which serves as the basis for the catalytic flame analysis. Over the Reynolds numbers (*Re*) investigated in the non-catalytic burner, no flame is observed for $\varphi \leq 0.16$, and flame extinction occurs at *Re* < 571 and *Re* < 381 for $\varphi = 0.18$ and 0.20, respectively. Moreover, a significant amount of unburned H₂ exits the burner in all cases. With the Pt catalyst coated on the walls, complete H₂ combustion is attained for 0.10 $\leq \varphi \leq 0.20$ where the contribution of gas phase (homogeneous) reaction increases with *Re*. Furthermore, radiation on the wall and at the inlet affects the combustion kinetics and flame temperature. Finally, NO_x emission is investigated under the same conditions and found to increase with equivalence ratio but has a negligible effect with the inflow Reynolds number.

2.1 Introduction

Combustion using a catalyst is a promising technique to stabilise a premixed flame with low NO_x emission used in a number of applications [50,87,117,118] for many years. Recently, research on this technique has been growing with an aim to mitigate greenhouse gas emissions in combustion of hydrogen [119] or hydrogen enriched syngas mixtures [104]. Moreover, hydrogen enriched fuels are now of great interest to replace the fossil fuels in

most gas-powered systems [120]. One example is a honeycomb monolith burner [58] consisting of multiple channels used in natural gas fired boiler or turbine applications. In practice, hydrogen non-catalytic combustion in such systems is not simple because of flame instabilities leading to incomplete combustion [121,122]. Pizza et al. [121] investigated flame instabilities in a mesoscale planar burner considering heated wall with a hydrogen/air equivalence ratio of 0.5, and varying inflow velocities from 0.003 to 11 m/s. They categorised the instabilities as mild, ignition/extinction, oscillatory, symmetric, and asymmetric based on the flame shapes depending on the inflow velocities. In another study [123], they also discussed the similar flame instabilities in a microscale planar burner where the same inflow conditions were used. A flame can be stable, unstable or blowout in a particular inflow velocity depending on the hydrogen lean composition, as investigated in the experimental work of Schefer et al. [124] in a multi nozzle premixed burner. With an aim to increase the hydrogen/air flame stability, Yang et al. [85] experimentally investigated a converging-diverging tube burner considering various equivalence ratio (0.6-2.2) and inflow velocity (3.4 to 42 m/s). They found that the flame shape gets thicker and longer with an increased velocity, and the flame stability limit follows an increasing-decreasing trend with the equivalence ratio. However, the tendency to flashback of comparatively rich H_2/Air mixtures limits the applicability of non-catalytic burner due to safety issues. H.Pers et al. [125] experimentally explored possible flashback initiation of H₂/Air laminar flame in a premixed burner and discussed the effects of reactants preheat and wall temperature on flashback. Although some efforts are made considering pin fin arrays [126] and a preheater conductor plate [103] inside the reactor to maintain H₂/Air flame, this is only feasible with high inflow velocity.

In contrast, catalytic aided hydrogen combustion provides a lower activation energy [77], a higher flame stability limit [76], and the potential to reduce NO_x emissions [127]. However, hydrogen catalytic combustion involves complex chemical interactions with solid substrate, resulting in a number of homogeneous (gas phase) and heterogeneous (surface) reactions with increasing temperature [128], and the behaviour of which and impact on NO_x emission are beyond understanding in most applications. Therefore, prior to practical implementation, a comprehensive investigation is required for fundamental knowledge of heterogeneous kinetics and their coupling with corresponding homogeneous kinetics. Many studies have aimed at investigating hydrogen hetero-/homogeneous combustion in a planar burner because of its design simplicity and operation. For example, Appel *et al* [129] experimentally investigated the hydrogen/air mixtures over platinum in a planar burner under fuel lean

stoichiometric condition (0.28-0.32), providing results of the onset of homogeneous ignition at different laminar inflow operations. In the same study they numerically studied several hetero-/homogeneous reaction schemes (four homogeneous and three heterogeneous) and addressed the effect of various combination of chemistry coupling. To observe the effect of catalytic reactivity (A_s) , Pizza *et al.* [108] numerically studied a 1 mm height microchannel with an equivalence ratio of 0.5. The results suggest that the catalyst loading controls the mode of combustion and an active catalyst is required to suppress undesirable flame. Similarly, Choi et al. [130] investigated a platinum catalytic micro planar combustor for a range of inflow conditions at stoichiometric H₂/Air mixtures. They found that the platinum catalyst concentration has no effect on reaction characteristics. However, the requirement of a catalyst depends on the specific fuel and catalyst type discussed in the previous study [108]. Again, H₂ combustion using a catalyst is influenced by the wall thermal condition [131]. With an aim to improve hydrogen conversion rate, Y. Zhang et al. [131] investigated catalytic microchannel by varying solid wall thermal conductivity and outer wall heat transfer coefficient. They showed that the conversion rate of H₂ is increased with lower wall thermal conductivity and heat transfer coefficient. The same team [132] also studied the self-ignition process of H₂/Air in microchannel catalytic considering radiation and convective heat dissipation. They showed that the self-ignition process is influenced by several factors which are dependent on temperature. Again, Sui et al. [66] studied combustion stability limits of hydrogen/air mixtures (equivalence ratio = 0.4) in platinum-coated microchannels by considering radiation heat transfer and solid heat conduction. They showed that lower solid thermal conductivity provides wider combustion stability and radiation heat loss towards inlet has a substantial effect on lowering wall temperature.

Again, H₂/Air premixed combustion in a catalytic planar burner has been carried out at elevated pressures. Mantzaras *et al.* [133] experimentally and numerically studied a hydrogen/air planar combustor with a pressure range of up to 10 bar and found homogeneous combustion suppression at high pressure (> 4 bar) due to produced water that acts as a third body efficient radical for terminating gas phase reactions. Moreover, Ghermay *et al.* [76] investigated the same burner with preheat to gain a better understanding of the pressure/ temperature dependence and catalytic chemistry coupling effect. Over the operating conditions considered, the results showed that the mass transport limited conversion of hydrogen and homogeneous combustion could be sustained at high pressure with preheating. Furthermore, the effect of hetero-/homogeneous chemistry of hydrogen/air combustion in planar burner was studied by considering catalytic segmentation [132,134], burner aspect

ratio [135,136], multiple channel [68], and addition of intermediate and final product on homogeneous combustion [137,138]. Besides, Zhang *et al.* [112] studied a planar model with platinum catalyst and obtained the critical range of equivalence ratio for transitioning from coupling chemistry reaction to pure catalytic/heterogeneous reaction. In another study [131], they investigated the effect of wall thermophysical conditions on hydrogen catalytic reaction and compared the performance in terms of the reaction efficiency and flame stability, which were favourable at higher wall thermal resistance. Similarly, the recent experimental work of Lu *et al.* [81] reported that the wall heat loss has a significant effect on catalytic reaction. Additionally, they studied the critical equivalence ratio of transformation of reaction type (pure catalytic and hetero-/homogeneous reaction) for an equivalence ratio range of 0.2-1.2. Hence, the mode (or type) of reaction inside a catalytic burner is highly influenced by the wall thermal conditions. However, an exploration is required to observe the effect of inflow velocity on transforming the reaction type (pure catalytic and hetero-/homogeneous reaction) inside a catalytic burner.

As discussed above, a catalytic combustor of planar type was of prime interest in most studies as it provides efficient interaction with fuel species as well as design flexibility in various applications. Again, the low flame temperatures in such system are vital for zero NO_x emission. For this, the ultra-lean mixtures are preferable, however, there are limited investigations on it. The underlying physics of flame at an ultra-lean condition is to be investigated to get a full understanding of NO_x reduction process. Moreover, wall thermal condition in a planar burner was considered convective heat loss or fixed temperature in previous studies but an adiabatic assumption is more practical for modelling a multi-channel honeycomb burner as heat is uniformly distributed in this case. Hence, the heat conduction in solid wall has not been considered in the present study. Furthermore, a flame stability analysis is necessary for this condition to observe the challenges prior to switching to catalytic approach.

The present work undertakes a numerical study on both non-catalytic and catalytic planar models for hydrogen/air combustion. The computations on two-dimensional laminar flow species transport model with multicomponent diffusion were carried out. Both homogeneous and heterogeneous kinetics were included in species transport modelling. A stability map as a function of Reynolds number for homogeneous combustion under fuel lean operation of equivalence ratio of 0.18 and 0.20 was obtained. The objectives were to assess the gas phase stability map and to delineate the catalytic approach for hydrogen/air combustion with NO_x emission over a range of equivalence ratio (0.10-0.20).

The article is organised as follows. First, the numerical model of planar burner is validated with experimental measurements. Then a stability map is obtained and discussed for homogeneous combustion in non-catalytic burner. Transition of chemistry coupling in catalytic burner, and their effects are analysed with species average mole fraction profiles, average temperature profiles and contour plots. Finally, NO_x emission chrematistics are discussed.

2.2 Computational modelling

Figure 2.1 shows a schematic of a planar burner having a dimension of h=7 mm between two parallel Pt coated plates. The chemically reacting gas flow through the burner is governed by the Navier-Stokes, energy and species transport equations which are solved considering steady-state, laminar and multicomponent flow assumptions. The equations solved are stated as follows:



Figure 2.1 Schematic of the catalytic burner geometry (L – burner length, h – gap between wall)

Continuity equation:
$$\nabla .(\rho \mathbf{u}) = 0$$
 (2.1)

where ρ is the density, **u** is the velocity vector.

Momentum equations:
$$\nabla .(\rho \mathbf{u}\mathbf{u}) = \nabla .\mu [\nabla \mathbf{u} + (\nabla \mathbf{u})^T - \frac{2}{3}(\nabla .\mathbf{u})I] - \nabla p$$
 (2.2)

where μ is the dynamic viscosity, p is the pressure, I is the unit tensor.

Energy equation:
$$\nabla . (\boldsymbol{u}(\rho E + p)) = \nabla . (\lambda \nabla T - \sum_{i=1}^{N_g} h_i \boldsymbol{J}_i + \boldsymbol{\tau}. \boldsymbol{u})$$
 (2.3)

where E, T, λ , h_i , J_i and τ denote the total energy including pressure work and kinetic energy, temperature, thermal conductivity, species enthalpy, species diffusion flux and viscous stress tensor, respectively. Here, N_g is the total number of gas species (*i*).

Species transport equations:
$$\nabla . (\rho \boldsymbol{u} Y_i) = -\nabla . \boldsymbol{J}_i + \dot{w}_i$$
 (2.4)

Here, Y_i is the species mass fraction. The species diffusion fluxes (J_i) are computed from the multicomponent diffusion equations of Maxwell-Stefan [139], considering the thermal diffusion [139] for light species. The source term, \dot{w}_i is the result of rate of production and destruction of gas species (i).

Surface species coverage equations:
$$\frac{d\theta_j}{dt} = \frac{\dot{s}_j}{\Gamma} = 0$$
 (*j*=1, 2,...., *N_s*) (2.5)

where θ , *s* and Γ are the surface species coverage, surface species molar production rate and surface site density, respectively. The alteration of species site coverage that is fraction of surface sites covered by species (*j*) calculated from the above equation. The production rate (*s*) for each surface species is computed considering both gas and surface species' production and destruction by surface reactions. Here, *m* is the total number of surface species (*j*). At the steady-state condition, the transient term of the equation vanishes and the net production rate equals to zero.

The boundary condition for burner wall is considered to be no slip and adiabatic. The interfacial gas-wall boundary (y=0 and y=h) conditions for gas-phase species are ($\rho Y_i V_{i,y}$) $_y=M_i \hat{s}_i$. Where V_i , M_i , and \hat{s}_i are the gaseous species diffusion velocity, molecular weight and heterogeneous molar production rate, respectively. Inflow boundary conditions are set uniform for velocity, temperature, and the species mass fraction. A zero gradient boundary condition is imposed at the outlet for all the properties and the pressure is specified as atmospheric. To simulate the radiation effects between the inner surfaces, at the inlet and outlet, the discrete ordinates (DO) model is used. The DO radiation heat transfer from a hot reaction zone towards the inlet and outlet are considered at boundary temperature. The internal emissivity of 1.0 is set for Pt-coated surface, the inlet and outlet enclosures. The Pt catalyst wall acts as an igniting medium of the incoming premixed H₂/air mixture, releasing radicals and heat that promote the gas phase reaction. H₂/air lean mixtures (varying $\varphi=0.10$ -0.20) are considered at the inlet temperature of 312 K. Such flow conditions are chosen to represent a range of low temperature heating applications, covering the inflow velocity from

0.075 m/s to 3.5 m/s at 1 atm. These conditions can be characterised by the flow Reynolds number based on the burner height (h=7 mm). The upper limit of the Reynolds number studied is 1333 which is below the critical Reynolds number 1400 and thus justifies the laminar assumption in the present configuration.

The above governing equations were discretised and solved on a structured grid using Finite Volume Method (FVM). The grid with 350×100 points (in the *x* axis and *y* axis, respectively) over the burner domain (250 mm \times 7 mm) was sufficient to obtain a grid independent solution. Grid towards the inlet and walls were refined to capture the high gradients of flow variables. The near upstream node and near wall node were positioned at x/h (from inlet) = 0.033 mm and y/h (from catalytic wall) = 0.0061 mm, respectively. An example of grid test is presented in Figure 2.2 which ensures the grid requirement for the Reynolds number from 57 to 1333. However, the burner length in this study was also









Figure 2.2 Grid test: Transverse profiles at three selected streamwise locations, (a) hydrogen mole fraction (X_{H2}) (b) Temperature (T), $\varphi=0.20$, $T_{in}=312$ K

extended up to 700 mm for capturing the entire reaction zone at higher Reynolds numbers of interest. As a result, a total of 900 \times 100 grid points in the extended domain was used keeping the same resolution near upstream and also at the near wall region. Ansys Fluent 2020 R2 version was used to carry out computations. A 2-D double precession planar steady-state solver was selected, and viscous model was set to laminar. Species transport model was used to solve the volumetric and wall surface reactions using a stiff chemistry solver with finite rate chemistry. For pressure velocity coupling, SIMPLE algorithm was used. Spatial discretization method for the gradients and pressure were specified as least square cell based and second order, respectively. To ensure accuracy, momentum, energy, and species equations were spatially discretised with a second order upwind method. The simulation terminates when the convergence criteria of 10⁻⁶ for all the residuals were met or the residuals approach steady states.

2.2.1 Chemical kinetics

A detailed mechanism of surface reactions [140] for hydrogen oxidation over the platinum catalyst, was used in this study. The mechanism consists of eleven irreversible and three reversible reactions and, has five surface and six gaseous species. The platinum surface site density (Γ) was set to 2.7×10⁻⁹ moles/cm² [140]. The homogeneous gas phase chemistry proposed by Warnatz et al. [141] was used with the heterogeneous surface chemistry, which includes nine species and nineteen elementary reactions. CHEMKIN [142] and Surface-CHEMKIN [143] were used to calculate the homogeneous and heterogeneous reaction rates, respectively, while the transport properties were evaluated from the CHEMKIN transport database [144]. Furthermore, to simulate NO_x emission, a NO_x kinetic scheme is added to gas phase kinetics for the catalytic burner. As mentioned in many studies, NO_x in H₂/air combustion comes mainly from NO and NO₂. NO can be produced in three ways: the thermal route using Zeldovich mechanism [145], the N₂O route and the NNH route [26]. The thermal NO is significant in high flame temperature combustion which is available in the Ansys Fluent module and estimated at post-processing stage. The kinetics of the N₂O route and NNH route are taken from Glarborg et al. [26] and are considered responsible for NO_x emission over a wide range of lean combustion. The well-established NO₂ reaction of Howard et al. [27], is considered to estimate the NO₂. However, the mixture gas viscosity and thermal conductivity were computed using mass weighted mixing law. Both the multicomponent and thermal diffusions were considered in the simulation and the kinetic theory was used for the calculation of binary mass diffusion coefficients and thermal diffusion coefficients.



(b)



Figure 2.3 Comparisons of (a) species and temperature profiles at axial positions, experiment [129] (b) OH contour plot (arrow indicates the onset of homogeneous ignition), $\varphi = 0.28$, $T_{in} = 312 \text{ K}, \text{ Re} = 762$

2.2.2 Validation of the numerical simulations

In order to validate the numerical model, the computed results are compared against the existing experimental results [129]. The burner configuration chosen was the same as experiment [129]. Inflow parameters from the experiment were copied exactly in simulation and the measured temperatures along the burner length were used as a wall boundary condition. An extensive validation was carried out at different flow conditions and, the profiles of species mole fraction as well as temperature in five axial locations were compared. An example of validated results for φ =0.28, T_{in} =312 K, Re =762 is shown in Figure 2.3(a). The overall agreement between the measured and computed results is excellent and the maximum deviation is estimated only less than 3%. Besides, a comparison of the onset of homogeneous ignition is also presented in terms of the OH contour plot in Figure 2.3(b) which further confirms the accuracy of the numerical model used in the work.

2.3 Results and discussion

A stability map of the fuel lean H_2 /air combustion in non-catalytic burner is firstly presented, to gain a comprehensive understanding of the challenges and also to facilitate the discussion of the importance of using a catalyst under similar operating conditions. Then, a comparison is made between pure catalytic and coupled chemistry (hetero-/homogeneous reactions). Finally, NO_x emissions are computed.

2.3.1 Flame stability map in non-catalytic burner

The flame stability map of premixed fuel lean H₂/air is shown in Figure 2.4(a). The computations of fuel lean conditions were performed under wall adiabatic condition by varying the inflow Reynolds number (*Re*). The φ for H₂/air is considered here just above the lower flammability limit from 0.15 to 0.20. Prior to combustion, the H₂/air mixtures is ignited on both surface wall at a distance 0.5*h* from inlet. The ignition temperature is set equal to adiabatic temperature of respective stoichiometric condition. The average outlet temperature (*T*_{out,avg}) (Left axis) of products and the dimensionless heat loss at the inlet (*Q*_{loss}/*HoR*_a) (1st right axis) and unburned H₂(%) (2nd right axis) exiting the burner outlet are presented as a function of *Re*. Here, the *Q*_{loss} is defined as the net heat leaving the burner through the inlet and calculated from the following expression:

$$Q_{loss} = Q_{rad} - H \tag{2.6}$$

Where Q_{rad} and H are the radiation heat and enthalpy flux through the inlet boundary, respectively. The Q_{loss} is made dimensionless with the actual heat of reaction (HoR_a) inside

the burner. The dashed and solid lines denote the results of the equivalence ratio (φ) of 0.18 and 0.20, respectively. It should be noted that for $\varphi \leq 0.16$ no flame is observed under the present operating conditions, but the flame extinction lower limit for $\varphi = 0.18$ and $\varphi = 0.20$ is found below Re < 571 and Re < 381, respectively as pointed out by the arrows in the temperature results. The $T_{out,avg}$ measured both cases gradually increases with Re and this variation is due to the unburned H₂ exiting the burner and the radiation heat loss at the inlet. At a low Re, high radiation heat loss causes a low flame temperature in the reaction zone and consequently, less H₂ conversion. The maximum unburned H₂ at the near flame extinction limit is obtained 11.5 % and 10.3 % for $\varphi = 0.18$ and $\varphi = 0.20$, respectively. Again, as the Re increases, the radiation heat loss gradually decreases. Therefore, there is increased flame temperature with Re. However, the flame extinction higher limit is not obtained within the Re studied here. Figure 2.4(b) shows average mole fraction (Left axis) of H₂ and H₂O, and average temperature (Right axis) for $\varphi = 0.20$ along the burner length (x/h). Solid line and dashed line denote the results of Re = 762 and Re = 1333, respectively. The OH mole fraction on the wall shows the onset position where the flames are ignited.

As shown in Figure 2.4(b), the flame ignition for both *Re* is almost at the same position and, precisely within 0.20-0.30 of x/h. The T_{avg} increases up to a certain burner length as a result of the high rate of H₂ consumption, then decreases to a constant value. As expected, the trend of $X_{H2O,avg}$ is similar. However, the position at which the flame peak temperature occurs is varied respectively at x/h=10 and x/h=14.3 for Re = 762 and Re = 1333. The interesting point is that there is no change in $X_{H2,avg}$ beyond the position of the flame peak temperature for both cases. The reason is the position of reaction zone attached to the wall keeping H_2 unburned in the burner centre region as shown in Figure 2.5(a). As the results indicate, most of H₂ is consumed within the reaction zone and after that no further reaction of H₂ takes place. Again, the peak temperatures exceed the adiabatic flame temperature ($T_{adb} = 937$ K) and are pronounced near the wall of the burner. This is because of diffusional imbalance of the H_2/air mixture as the Lewis number of H_2 is below unity (i.e. *Le*=0.3). Therefore, the H_2 species high affinity towards the hot reaction zone causes a comparatively rich mixture and high temperature. To get a better understanding of the reaction zone, the temperature contour plots are shown in Figure 2.5(b) for three different *Re*. As the main interest is on the reaction zone near upstream, only 3/7 of the burner length is presented. The high temperature region near the wall is the reaction zone and the shape of the zone is symmetric about the burner mid plane. At low Re = 381, the reaction zone is confined to a small region near upstream. As *Re* increases, the reaction zone elongates along the

burner length. Such behaviour was also predicted in the same burner by Pizza *et al.* [121] but at different inflow conditions ($\varphi = 0.50$ and 1110 < Re < 2960), and they defined the flame as an open symmetric steady flame. However, it is worth noting that complete H₂ conversion is not attained at the burner exit under the current operating condition.





Figure 2.4 H₂/air combustion in non-catalytic burner (a) stability map as a function *Re* (b) Average value along the burner length ($\varphi = 0.20$, $T_{in} = 312$ K): Left Axis- Mole fraction (X_{avg}); Right Axis- Temperature (T_{avg}). OH mole fraction is taken on surface wall. *Re*=762 (solid line); *Re*=1333 (dashed line)



Figure 2.5 Contour plots at different Reynolds numbers inside non-catalytic burner for φ =0.20, T_{in} =312 K (a) hydrogen mole fraction (X_{H2}) (b) Temperature (T)

2.3.2 Catalytic burner combustion

To investigate the catalytic effect in the same burner, the wall is considered as a Pt coated reacting surface, and the elementary chemistry of Deutchmann *et al.* [140] is implemented including the gas phase chemistry. The other boundary conditions are kept similar as in the non-catalytic burner.

2.3.2.1 Flame stability

Figure 2.6 shows the OH contours inside the catalytic burner at different operating conditions. The results are presented for four equivalence ratios (φ) of 0.10, 0.15, 0.18 and 0.20. H₂ conversion is achieved 99.9% at the burner exit almost in all the cases except in a few cases where φ = 0.10 at high Reynolds number. Only the X_{OH} contour plot upto x/h = 30 is shown to focus the interest. The X_{OH} scale is kept 0-10⁻⁴ to capture the onset of homogeneous combustion. The equivalence ratio of φ = 0.10 represents an ultra-lean mixture as that is below the lower flammability limit of hydrogen. For this case, no significant X_{OH} is observed over the range of *Re* considered, which indicates pure catalytic conversion of H₂. At a near flammability limit (φ = 0.15), the flame is ignited near the burner wall at a position between x/h= 5 to x/h= 7.5 when the *Re* is 571.



Figure 2.6 OH contour plot inside catalytic burner (up to x/h = 30) at different *Re*, $T_{in}=312$ K

However, as the *Re* increases, the onset position remains approximately the same, and the flame shape is elongated along the burner length. Whereas, at a lower Re < 571, flame is not ignited because of the high radiation heat loss at the inlet as shown in Figure 2.7(a). Under such conditions, pure catalytic (i.e., heterogeneous) chemistry (PC) plays the main role in complete H₂ combustion, which will be discussed in more details in the later section. Again, flame ignition occurs at a lower Re = 381 for $\varphi = 0.18$ and the onset position shifts toward the inlet. This is due to the comparatively rich H₂/air mixture closer to the upstream near catalytic surface and releases high heat promoting flame ignition. This effect is also observed for $\varphi = 0.20$ where the onset position is very close to the inlet.

Figure 2.7(a) shows the dimensionless heat loss (Q_{loss}/HoR_a) (right axis) at the inlet and the convective heat (*H*) (left axis) as a function of *Re*. The heat loss is dependent on φ as the radiating temperatures are different. At a low *Re* for all φ , the heat loss is very high and then gradually decreases with *Re*. However, the heat loss becomes negative at high *Re* > 952 cases for $\varphi = 0.10$. This is because the *H* dominates over Q_{rad} at the inlet, and thus the ($Q_{rad} - H$) becomes negative which further indicates no heat leaving the burner inlet.



Figure 2.7 Catalytic burner: (a) Left axis: Enthalpy flux at inlet (*H*), Right axis: Net heat loss at inlet (Q_{loss}/HoR_a) (b) Average outlet temperature ($T_{avg,out}$), T_{in} =312 K

As expected, the convective heat increases with Re and is independent on φ . As mentioned earlier, the radiation heat loss through inlet has significant influence on the flame temperature and reaction kinetics. However, the average flame outlet temperatures ($T_{out,avg}$) with increasing Re for catalytic combustion are shown in Figure 2.7(b). The arrow denotes the transition of pure catalytic chemistry (PC) to coupled (hetero-/homogeneous) chemistry (CC) reaction.

2.3.2.2 Effect of coupled (hetero-/homogeneous) chemistry

The distribution of species mole fraction and temperature on the catalytic wall are shown in Figure 2.8 for $\varphi = 0.20$. To identify the effect of the homogeneous chemistry on catalytic

combustion, two Reynolds numbers, 190 and 762, are selected for comparison where the pure catalytic chemistry (PC) and coupled chemistry (CC) are dominant, respectively. At the pure catalytic mode, the computation is performed without considering the homogeneous chemistry. Figure 2.8(a) shows a comparison of the results of PC (dashed line) and CC (solid line) for Re= 190. There is no variation observed in the species mole fraction of H₂, H₂O and OH and wall temperature (T_W). The catalytic conversion starts at the beginning of upstream wall resulting in a high temperature near upstream close to the wall. Then, it decreases as there is gradual reduction of H₂ is left.

Similarly, no changes between the two chemistries are found in the surface coverages as shown in Figure 2.9(a), where the PT(s) and O(s) constitute the main coverage. Here, the surface coverage is the fraction of surface sites covered by species and PT(s) is available surface sites for adsorption. O₂ adsorption/desorption has strong dependence on the wall temperature as discussed in the earlier studies [80,140]. As the temperature decreases, the excess O_2 allows for O_2 adsorption that consequently promotes H_2 reaction. Therefore, H₂ is completely consumed. The available uncovered surface sites PT(s) are then used for further O₂ adsorption. However, all the results indicate that the contribution of the homogeneous chemistry on the species concentration is negligible for Re=190. By comparison, the effect of homogeneous chemistry is significant for *Re*=762 shown in Figure 2.8(b). The X_{H2} distribution on the wall for PC decreases gradually but the distribution for CC is very low and, approaches zero within a certain burner length. This is due to the homogeneous reaction zone in proximity to the catalytic wall. Therefore, most of H₂ near the surface is burned in the reaction zone, reducing the H₂ level on the catalytic surface. Consequently, the H₂O wall distribution for CC is found to be higher. However, the X_{OH} distribution on the wall is lower for CC because of the strong influence of the catalytic reaction.

Previous studies [69,129] showed that the catalytic reaction influences the homogeneous ignition as the catalyst itself produces less OH. So, catalyst is an efficient sink of homogeneously produced OH inhibiting homogeneous reaction. Moreover there is significant variation of wall temperatures between two chemistry (PC and CC) which is discussed in later section. A comparison of the surface coverages for Re = 762 is shown in Figure 2.9(b). As expected, there is a large variation in the surface coverage values near the homogeneous reaction zones. The hydrogen adsorption H(s) for CC is found to be less as there is high H₂ consumption in the reaction zone. For example, the maximum

variation between two chemistry (CC and PC) is at a distance x/h = 7.7 from inlet where temperature difference is maximum. Here, the H(s) (×10⁴) level for CC and PC are 0.11and 0.48, respectively. Again, the OH adsorption in CC is higher due to the influence of the catalytic reaction. At x/h = 7.7, the OH(s) (×50) level for CC and PC are 0.202 and 0.141, respectively. However, there is no significant variation observed in H₂O(s) at that position.



Figure 2.8 Comparison of species mole fraction and temperature distribution on wall between Coupled (CC) and pure catalytic (PC) chemistry, $\varphi=0.20$, $T_{in}=312$ K (a) Re=190, (b) Re=762



Figure 2.9 Comparison of species surface coverage on wall between Coupled (CC) and pure catalytic (PC) chemistry, φ =0.20, T_{in} =312 K (a) Re =190, (b) Re =762

2.3.2.3 Surface temperatures

The maximum surface temperature, T_W , in all cases depicted in Figure 2.10, exceeds the adiabatic flame temperature. This is due to the diffusional characteristics of the H₂/air mixture which can be described by the theoretical wall temperature ($T_{w,max,th}$). In the case of the catalytic surface reaction on a flat plate, it is shown in [146] that $T_{w,max,th}$ remains constant along the plate under adiabatic conditions and is determined as:

$$T_{w,\max,th} = T_{\infty} + Le_{H_{2}}^{-2/3}(\Delta T)$$
(2.7)

where Le_{H_2} is the hydrogen Lewis number and its value of 0.30 is considered in all the cases, T_{∞} is the free stream temperature, and ΔT is the combustion temperature rise from the adiabatic temperature:

$$\Delta T = T_{adb} - T_{\infty} \equiv Y_{H_2,\infty} Q_{H_2} / C_p \tag{2.8}$$



Figure 2.10 Maximum wall temperature using pure catalytic chemistry (PC), T_{in} =312 K

Here, T_{adb} is the adiabatic flame temperature, Y_{H_2} is the free stream hydrogen mass fraction, Q_{H_2} is the combustion heat per unit mass of hydrogen. At a unity Lewis number, the H₂/air flame temperature is identical to the adiabatic temperature but it always becomes superadiabatic temperature ($T_{w,max,th} > T_{adb}$) at a Lewis number less than unity. Figure 2.10 shows the maximum wall temperature using the pure catalytic chemistry (PC) for $0.10 \le \phi \le 0.20$ along with $T_{w,max,th}$ calculated from equation (2.7). The computed maximum wall temperatures ($T_{W,max}$) for the Reynolds numbers of 190 and 762 are shown, considering with and without radiation. The $T_{W,max}$ values are identical to $T_{w,max,th}$ irrespective of the Reynolds number when the radiation model is not included. With radiation, the predicted catalytic peak temperatures are lower than the theoretical value due to the radiative heat loss towards the inlet. In particular, at a lower Re= 190, the $T_{W,max}$ are comparatively very low because of the high heat losses at the inlet discussed in Figure 2.7. However, the wall temperature for CC in Figure 2.8 is lower than PC in the region of homogeneous reaction zone. This is because of the shielding effect of the homogeneous reaction zone that limits the surface superadiabaticity induced by the catalyst [133]. Furthermore, there are up/down peaks on the wall temperature near inlet as shown in Figure 2.8. These peaks are mainly from the effect of radiation. To observe this effect, the incident radiation (G) and net radiation heat flux (q_{rad}) at the inlet and on the wall surface are presented in Figure 2.11. At the inlet, Figure 2.11(a), the incident radiation is maximum at the wall and then gradually deceases to a minimum at midplane for Re=762. For *Re*=190, the minimum occurs at $y/h \approx 0.15$ at a distance from bottom wall (which is the same in case of top wall because of symmetry) and then reaches to peak at $y/h \approx$ 0.20. Apart from that there is no significant variation. The magnitude between two Re differs significantly as the amount of fuel burned is different. However, the radiation heat fluxes have significant variation within $y/h \approx 0.20$ from the bottom wall for both cases (Re=190 and 762). Again, the trend and magnitude are approximately similar. The plot for Re = 762 shifted toward wall is expected as the net heat loss for both cases are not same. Here, it should be noted that the negative heat flux represents the radiative heat flux leaving the burner whereas the positive value is the opposite.

However, the up/down peaks of incident radiation are observed on surface wall near upstream within the burner length (x/h) of 5 for both cases. This is the effect that causes the up/down peaks of the wall temperature near upstream (Figure 2.8Figure 2.8) and consequently affects the reaction kinetics on the catalytic wall (Figure 2.9). After x/h > 5, the incident radiation decreases gradually without showing any variation. Consequently, the radiation heat fluxes are maximum at inlet and then decrease along the length. This becomes negative within the range 5 ~20 and 15 ~40 of x/h for Re = 190 and Re = 762, respectively. This further indicates that the radiation flux directed towards the wall in that region and then, it becomes zero because of the uniform flame temperature.



Figure 2.11 Variation of Incident Radiation (Left Axis) and Net Radiation Flux (Right axis), Coupled chemistry (CC), φ =0.20, T_{in} =312 K at (a) inlet (b) wall

2.3.2.4 H₂ conversion

The percentage of H₂ converted for $\varphi = 0.20$ at different *Re* is shown in Figure 2.12(a) along the length of the burner. The percentage is estimated from the average results at different axial positions of the burner:

$$H_{2}Conversion(\%) = \frac{Y_{H2,avg,in} - Y_{H2,avg,x}}{Y_{H2,avg,in}} \times 100$$
(2.9)



Figure 2.12 (a) H₂ conversion at different *Re*, (b) H₂ and H₂O distribution at x/h = 0.5, Coupled chemistry (CC), $\varphi=0.20$, $T_{in}=312$ K

Where $Y_{H2,avg,in}$ is the average H₂ mole fraction at the inlet, $Y_{H2,avg,x}$ is the average H₂ mole fraction at axial positions. For all cases, almost 99.9 % H₂ conversion is achieved within the burner length considered in this study. By comparison, the complete H₂ conversion for lower *Re* is obtained at shorter length because of the higher residence time shown in Figure 2.13. The residence time is calculated from the minimum burner length (L_c) for 99.9% H₂ conversion and inflow velocity. For $\varphi = 0.20$, the relation $L_c = 0.0005 \ Re + 0.025$ is obtained. However, the zoomed view in Figure 2.12(a) shows a significant catalytic conversion near upstream. For Re = 57, the conversion is above 22% at near upstream and, this becomes comparatively less at higher Re. The main reason is a high rate of H₂ consumption in the entrance region of the burner. Again, the low inflow and high residence time at a lower Re facilitates H₂ diffusion towards the catalytic surface. For clarification, Figure 2.12(b) shows the mole fraction distribution of H₂ and H₂O at x/h=0.5 from inlet. The vertical up and down arrows denote X_{H2O} (dashed) and X_{H2} (solid), respectively. The low H₂ level near wall for all cases indicates that catalytic reaction is practically mass transport limited. However, the H₂ distribution across the burner for Re = 57 is low because of the strong H₂ diffusion to the catalytic wall. Consequently, this produces high H₂O (dashedblack). When *Re* increases, the flow becomes convectively dominant and thus reduces the residence time for H₂ diffusion. As a result, the H₂O production is comparatively less with increasing Re.



Figure 2.13 Residence Time as a function of *Re* using coupled chemistry (CC), φ =0.20, *T_{in}*=312 K

However, H_2 transport toward the catalytic surface is governed by the catalytic reaction, which depletes both H_2 gas and surface species and maintains a mass transfer between two species. In reality, the species boundary layer is developed on the catalytic surface that inhibits the mass transfer. Therefore, the mass transfer coefficient can be used as a measure of resistance to mass transfer between the mean species composition and the composition at the reacting surface. This is a useful approach if the mass transfer coefficient is calculated quantitatively but, for a complex catalytic reacting flow, the mass transfer coefficient cannot be correlated in a simple way. Nevertheless, they can be used qualitatively to predict the catalytic reacting flow using the dimensionless Sherwood number (*Sh*).

$$Sh = \frac{k_c h}{D_m} \tag{2.10}$$

Where k_c is the mass transfer coefficient and D_m is the mass diffusivity for fuel species (H₂). The expression for k_c is as follows:

$$k_{c} = \frac{D_{m} \frac{\delta C_{H2}}{\delta y} \Big|_{y=0}}{C_{H2,W} - C_{H2,m}}$$
(2.11)

As noted in the above equation, $\delta C_{H2}/\delta y$ is the gradient of the species molar concentration at surface. The $C_{H2,W}$ and $C_{H2,m}$ are the surface and the mean concentration of H₂, respectively. The $C_{H2,m}$ is transversely calculated from the following equation.

$$C_{H2,m} = \frac{\int_{0}^{n} C_{H2} u dy}{\int_{0}^{h} u dy}$$
(2.12)

In this equation, *h* is the burner height, *u* is the local velocity and C_{H2} is the local molar concentration of H₂. Figure 2.14 shows the computed axial Sherwood number of H₂ species for three different *Re*. As expected in all the cases, *Sh* have high initial values near upstream, which are associated with the very thin H₂ boundary layer developed at the leading edge of the catalyst. This sharply decreases to a minimum value ~ 3.0 at x/h = 2.0 for *Re*=190, and then, increases to an asymptotic value of around 7.6 at x/h = 20, indicating no significant catalytic activity beyond this length. The behaviour of sharp decrement in the near inlet region is due to the high temperature differences between the wall and bulk temperature, which significantly affects the property of light species like H₂. Previous studies [147]



Figure 2.14 Local Sherwood Number (*Sh*) along the burner length for H₂, Coupled chemistry (CC), φ =0.20, T_{in} =312 K

showed such effect of temperature on the H₂ properties and other dimensionless quantities inside a pipe for non-reacting flow.

2.3.2.5 NO_x emission

The NO_x emission for ultra-lean condition is calculated in present catalytic burner keeping the same boundary condition. The inclusion of NO_x kinetics into gas phase kinetics in the present study did not affect the concentration of other species or temperatures. To justify the NO_x model, experimental data of Anderson *et al.* [148] is used for comparison. Though the experiment was done at very high inflow velocity (15-18 m/s) and high pressure (3.8 and 5.2 atm) in a cylindrical burner (0.103 in diameter and 0.31 m long), the NO_x results reported were only dependent on the high flame temperature and fuel compositions. Therefore, NO_x values at a reasonable operating condition in present catalytic burner are compared. Over the equivalence ratios considered in Figure 2.15(a), the NO_x model shows a good agreement with the experiment. The contribution of NO₂ in total emission is computed less than 0.5 %in all cases. Thermal NO in Figure 2.15(b) is evaluated at post-processing stage at the same operating condition without considering radiation. The thermal NO obtained 5×10^{-5} ppmv and 0.041 ppmv for $\varphi=0.25$ ($T_{adb}=1327$ K) and $\varphi=0.39$ ($T_{adb}=1658$ K), respectively. Again, the total calculated NO_x for φ =0.25 and φ =0.39 are 0.09 ppmv and 0.57 ppmv, respectively. By comparison, thermal NO contribution is much less. However, NO_x emission at very lean condition within the range of $0.10 \le \varphi \le 0.20$ are computed for two Reynolds number of 762 and 1333 shown in Figure 2.16. The NO_x values at Re = 762 for $\varphi = 0.10, 0.13, 0.15, 0.18$ and 0.20 are 9.88×10^{-10} ppmv, 2.53×10^{-4} ppmv, 2.58×10^{-3} ppmv, 0.007 ppmv, and 0.016 ppmv, respectively. For Re = 1333, the NOx values have no significant variation for $0.10 \le \varphi \le 0.15$ but differ by approximately 0.002 ppmv for $\varphi = 0.18$ and 0.20. Furthermore, the NO_x values are comparatively less if radiation loss is considered.

(a)







Figure 2.15 Emission as a function of φ using coupled chemistry (CC) (a) NO_x, exp (Anderson et al. [148]) (b) Thermal NO, Re = 1333, $T_{in} = 600$ K



Figure 2.16 NO_x Emission as a function of φ using coupled chemistry (CC), $T_{in} = 312$ K

2.4 Conclusions

Hydrogen/air combustion in a planar burner with platinum coating along its length was numerically investigated. Computations were carried out in multicomponent species transport model of laminar solver using Ansys Fluent. Prior to analysis, the model was validated with available experimental results. To reduce NO_x emission, hydrogen/air lean conditions of equivalence ratio from 0.10 to 0.20 were considered for computation. The key findings of this work are:

- Homogeneous combustion over the range of equivalence ratios from 0.15 to 0.20 in the non-catalytic burner results in an incomplete combustion causing the flame temperature less than the adiabatic temperature. Flame extinction occurs below *Re* < 381 and *Re* < 571 for φ =0.20 and 0.18, respectively. No flame is observed for φ ≤ 0.16 over the *Re* considered.
- In the catalytic configuration, almost complete H₂ conversion is obtained for 0.1≤ φ ≤ 0.20 and both combustion modes (PC/CC) are influenced by the inflow Reynolds number (*Re*). With φ, the onset of flame ignition shifts toward the inlet.
- The radiation loss at the burner inlet has a significant effect on lowering the flame temperature, which also affects the combustion kinetics.
- Under the wall thermal condition analysed, sufficient catalytic burner length required for complete combustion is influenced by the inflow Reynolds number and residence time.

 NO_x emissions were controlled by N₂O and NNH routes compared to thermal NO. The NO_x values in the catalytic burner increase with φ and can be considered independent on the Reynolds number.

However, the amount of catalyst needed for the catalytic process in the planar burner varies significantly with different operating conditions, which poses a major challenge for its broader application. Therefore, modifications to the burner design and the effects of various catalyst coatings are explored in the next chapter.

Chapter 3: Enhancing the performance of catalysts in turbulent premixed fuel-lean hydrogen/air combustion

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Abstract

Catalytic-aided combustion is a proven technique for burning highly lean and ultra-lean mixtures of hydrogen and air. However, the noble catalyst required for combustion is naturally scarce and therefore expensive. In this study, we focus on a numerical investigation to determine the best way of coating a platinum catalyst inside a catalytic hydrogen reactor. We study various planar and non-planar reactors and find that the reactor with a combination of half and full cylinders is the most effective in H₂ conversion. Compared to an equivalent catalytic planar reactor, the non-planar configuration increases the H₂ conversion by 30.7%. The results show that enhancing mass and heat convection can significantly increase the H₂ conversion. Furthermore, in a non-planar reactor, surfaces with enhanced mass and heat transfer can achieve up to 50% catalyst savings when coated with a catalyst, while still maintaining a conversion rate of 2 kg/s per unit of catalytically-coated surface area.

3.1 Introduction

Catalytic Stabilised Combustion (CSC) is an effective approach for enhancing the fuel conversion and minimising NO_x emissions in a fuel-lean mixture combustion system [50,70,127]. This method has been applied in a variety of combustion applications, ranging from a portable micro-scale [108,123,134] to a large-scale [75,102,149] heat and power generation system. However, CSC processes in these systems involve a complex set of reaction mechanisms, that includes both catalytic (heterogeneous) and gas-phase (homogeneous) reactions. In a recent work [150], the essential physico-chemicals behind a CSC process were investigated. It was found that the use of a catalyst initiates the surface catalytic reaction, consuming a portion of fuel and subsequently releasing gas-phase

molecules. These molecules then promote the gas-phase reactions and further consume the remaining unburned fuel. However, the gas-phase reaction is largely dependent on the fuel concentration, inflow mass flow rate, preheat temperature, and pressure, as reported in the other articles [66,69,76,104,146]. Importantly, for a very lean mixture at atmospheric pressure, the gas-phase reaction occurs near the combustor wall and becomes dominant at a temperature above 1200 K [129]. In contrast, the fuel conversion at a temperature below 1200 K occurs entirely through the catalytic route which however strongly depends on the amount of the catalyst-coated surface used in combustor. Nevertheless, the catalytic surface reactions are very fast and mass transport-limited [140,150], which limits the fuel conversion. This requires further research for a fundamental understanding of the catalytic process to enhance the catalyst-substrate interaction.

However, a widespread use of the CSC process also comes with a major challenge due to its high cost and limited availability of catalysts, particularly noble metals. Therefore, extensive research is required to identify the most suitable catalyst that can deliver a high catalytic performance. To enhance the catalytic fuel conversion, several geometric modifications have been proposed, including increasing the surface-to-volume (S/V) ratio of a planar reactor. Zade *et al.* [109] studied this approach in a planar catalytic reactor with a H₂/Air mixture. They found that the H₂ diffusive flux to the reactor walls and inhibition of the gas-phase reaction increase with the S/V ratio. Similarly, Ghermay et al. [76] investigated both planar and tubular reactors with the characteristic lengths varying from 0.5 mm to 2.0 mm. Under a turbine-like operating condition with a large confinement (S/V), they showed that the reactor inhibits gas-phase combustion at an atmospheric pressure with a surface temperature of up to 1350 K and a preheat temperature of up to 773 K. This approach improves the reactor catalytic efficiency, thus reducing the catalytic surface length and the amount of catalyst used. In addition to increasing the S/V ratio, wall deformation, such as cavities, obstacles, and waviness, can also improve the catalytic performance. For instance, introducing cavities in the design of a micro-reactor has been shown to enhance mass and heat transfer, resulting in improved catalytic conversion, as reported by Li *et al.* [151,152]. They showed that the cavities allow better mixing of the reactants to enhance mass and heat transfer, thus resulting in an improved catalytic conversion.

Similarly, Chababe *et al.* [107] numerically delineated the effects of both cavities and obstacles with a multi-segment coating. They obtained an optimised configuration using obstacles with segmented catalytic walls, which are capable of high catalytic conversion for flame stabilisation. The obstacle walls were coated; thus, these regimes have an effective

high catalytic S/V ratio that favours the catalytic reaction. Both Hunt *et al.* [106] and Esfandiary *et al.* [105] used surface waviness on the reactor walls considering both continuous and discrete coatings. The function of surface waviness is to interrupt the flow and modify the boundary layer of reactants, which alters the catalytic activity. They optimised the reactor configuration with discrete coating and improved the catalytic performance by up to 400% and 459% using platinum and nickel, respectively. Discrete coatings were chosen on the wavy surfaces where the Nusselt and Sherwood numbers were higher. Overall, these modifications improved the catalytic efficiency, reduced the catalytic surface length, and used less catalyst. They achieved this by increasing the S/V ratio, which plays a key role in enhancing mass transfer towards the catalytic surface and improving catalytic reactivity.

of Nonetheless, studies hydrogen/air catalytic combustion most [87,101,102,119,129,133,153–155], either experimental or numerical, are limited to the reactors of planar configurations. As discussed earlier, a detailed understanding of the underlying physics inside a reactor with surface deformations is a key path towards improving the performance of catalytic hydrogen/air combustion as well as optimising the use of catalyst. Again, the size of a catalytic planar reactor is crucial for achieving complete combustion of a lean hydrogen/air mixture. Also, from an economic standpoint, an excessive use of catalyst with the catalytic reactor length longer than required is not useful. So, compactness in the design of a catalytic reactor is required for the optimum use of the catalyst for hydrogen conversion. Moreover, the catalytic reacting flow in such cases becomes complex due to the turbulence generated by the flow. Therefore, this requires a thorough understanding of the catalytic process, which is equally important for both planar and nonplanar reactor configurations, before finding any potential improvements in CSC systems. For that purpose, a numerical simulation is a convenient and useful approach prior to an experimental investigation. Consequently, this study is focused on the numerical investigation of turbulent catalytic combustion of a premixed lean H₂/air mixture in the proposed configurations (planar and non-planar) to optimise the use of the catalyst for a compact reactor design. These configurations have been chosen with the practical use of a honeycomb burner in mind, where the heat transfer in solid materials can have a significant impact on the catalytic performance [68]. As a result, heat transfer in solid walls has been included in all configurations. Particular objectives are to provide a better understanding of the catalytic combustion process in the different configurations proposed, which leads to the investigation of catalyst optimisation.

This article is structured in the following manner: Section 3.2 presents the numerical methodology and validated results. Next, in Section 3.3, the effect of the S/V ratio among the different configurations is discussed. The best possible configurations are compared as a function of the catalytic conversion rate and Nusselt and Sherwood numbers in Section 3.4. Finally, conclusions are made in Section 3.5.

3.2 Modelling of turbulent catalytic reacting flow

In our previous study [150], we used a fuel-lean premixed H_2/air mixture at an equivalence ratio of $0.10 \le \varphi \le 0.20$ under laminar flow conditions. This mixture was chosen because of its suitability for a low-temperature heating application. Hence, a value of $\varphi = 0.15$ is selected in this work, which is within the studied fuel-lean equivalence ratio limit. Again, the inflow Reynolds number for laminar flow was considered up to 2666 based on the hydraulic diameter (2h) [150]. Therefore, to investigate the catalytic effect at a high Reynolds number in a turbulent regime, we have taken an inflow Reynold number ≥ 4200 for a planar reactor. For the reactor configurations with a confined cylinder, the Reynolds number is kept above 3385 (\approx 846 based on cylinder diameter). However, in both reactor configurations, the Reynolds numbers considered are above the critical Reynolds number: 3500~4000 for the planar channel [156], and 150~225 (based on cylinder diameter) for the channel with cylinder [32,33], which justifies the turbulent assumption in this study. Numerical computations are carried out with a RANS code that includes a species transport model and a detailed description of heterogeneous (surface) and homogenous (gas phase) kinetics. The surface mechanism of Deutschmann et al. [140] is used for modelling the catalytic reactions as it has been extensively validated against the experiments. For the gas phase reactions, five well-known mechanisms were tested and compared with measurements to find a suitable mechanism for predictions of the combined effect of the couple chemistry in the catalytic process. The governing equations and numerical settings to simulate the turbulent catalytic reacting flow are provided below.

3.2.1 Governing equations

Reynolds-Averaged Navier Stokes (RANS) computations with detailed chemistry (both homogeneous and heterogeneous) are performed to simulate turbulent reacting flow in a catalytic reactor with inner surfaces coated with Pt. The Navier-Stokes, species transport, and energy equations are solved considering Newtonian, incompressible, steady-state, and multicomponent flow assumptions. The equations are stated with the tensor notation as follows:

Continuity:
$$\frac{\partial}{\partial x_i} (\rho U_i) = 0,$$
 (3.13)

where ρ is the density, u=U+u' is the instantaneous velocity. U and u' are the mean and fluctuation components, respectively.

Momentum:

$$\frac{\partial}{\partial x_{j}} \left(\rho U_{i} U_{j} \right) = -\frac{\partial P}{\partial x_{i}} + \frac{\partial}{\partial x_{j}} \left[\tau_{ij} - \rho \overline{u_{i} u_{j}} \right], \qquad (3.14)$$

where $\rho u_i u_j$ is the Reynolds stresses and τ_{ij} is the viscous (or deviatoric) stress tensor,

$$\tau_{ij} = \mu \left(\frac{\partial U_i}{\partial x_j} + \frac{\partial U_j}{\partial x_i} - \frac{2}{3} \delta_{ij} \frac{\partial U_k}{\partial x_k} \right).$$
(3.15)

To model the Reynolds stresses, the low Reynolds (LR) variant of k- ε turbulence models proposed by Abe *et al.* [157] is chosen. This choice is due to the intense heating from the hot catalytic wall, which laminarises the turbulent flow to a certain extent, necessitating an overdissipative model compared to the standard k- ε model [102]. The overdissipation of turbulence is accomplished via the functions f_{μ} and f_2 in Equations (3.19) and (3.20). The LR model was validated for catalytic turbulent reacting flow [102], is therefore employed in this study. The equations used for the LR model are given below:

Turbulent kinetic energy (k):

$$\frac{\partial}{\partial x_{j}} \left(\rho U_{j} k \right) = \frac{\partial}{\partial x_{j}} \left[\left(\mu + \frac{\mu_{i}}{\sigma_{k}} \right) \frac{\partial k}{\partial x_{j}} \right] - \rho \overline{u_{i} u_{j}} \frac{\partial U_{i}}{\partial x_{j}} - \rho \varepsilon$$
(3.16)

Dissipation rate of k (ϵ):

$$\frac{\partial}{\partial x_{j}} \left(\rho U_{j} \varepsilon \right) = \frac{\partial}{\partial x_{j}} \left[\left(\mu + \frac{\mu_{t}}{\sigma_{\varepsilon}} \right) \frac{\partial \varepsilon}{\partial x_{j}} \right] - \rho C_{\varepsilon 1} \frac{\varepsilon}{k} \overline{u_{i} u_{j}} \frac{\partial U_{i}}{\partial x_{j}} - \rho C_{\varepsilon 2} f_{2} \frac{\varepsilon}{k} \varepsilon , \quad (3.17)$$

where

$$-\rho \overline{u_i u_j} = \mu_t \left(\frac{\partial U_i}{\partial x_j} + \frac{\partial U_j}{\partial x_i} \right) - \frac{2}{3} \rho k \delta_{ij} \quad \text{and} \quad \mu_t = \rho C_\mu f_\mu \frac{k^2}{\varepsilon}$$
(3.18)

In above equations, the model functions and constants are taken as the same as in the study of Abe *et al.* [157]: $\sigma_k=1.4$, $\sigma_{\varepsilon}=1.4$, $C_{\varepsilon l}=1.5$, $C_{\varepsilon 2}=1.9$, $C_{\mu}=0.09$,

$$f_{\mu} = \left[1 - \exp\left(-\frac{y^{*}}{14}\right)\right] \left[1 + \left(1 + \frac{5}{R_{t}^{3/4}} \exp\left\{-\left(\frac{R_{t}}{200}\right)^{2}\right\}\right)\right] \text{ and } (3.19)$$

$$f_{2} = \left[1 - 0.3 \exp\left\{-\left(\frac{R_{t}}{6.5}\right)^{2}\right\}\right] \left[1 - \exp\left(-\frac{y^{*}}{3.1}\right)\right]^{2}, \qquad (3.20)$$

where $y^*=u_{\varepsilon}y/v$, $R_t=k^2/v\varepsilon$ and $u_{\varepsilon}=(v\varepsilon)^{1/4}$ is the Kolmogorov velocity scale. The purpose of both the functions f_{μ} and f_2 is to make the model overdissipative compared to the standard high Reynolds number model, with both the functions approaching unity far from the wall [102].

Energy:
$$\frac{\partial}{\partial x_i} \left(u_i \left(\rho E + p \right) \right) = \frac{\partial}{\partial x_i} \left[\left(\lambda + \frac{c_p \mu_t}{\Pr_t} \right) \frac{\partial T}{\partial x_i} - \sum_{j=1}^n h_{g,j} J_{g,j} + \tau_{ij} u_i \right] + S_h, \quad (3.21)$$

where *E*, *T*, λ , *h*, *J* and *S*_h denote the total energy, temperature, effective conductivity, enthalpy, diffusion flux of species *j*, where *j* =1, 2, 3,....*N*_g, and sources of energy due to chemical reaction, respectively. *Pr*_t is the turbulent Prandtl number, and its value is set to 0.85.

Species transport:
$$\frac{\partial}{\partial x_i} \left(\rho u_i Y_{g,j} \right) = \frac{\mu_t}{Sc_t} \frac{\partial Y_{g,j}}{\partial x_i} - \frac{\partial J_{g,j}}{\partial x_i} + R_{g,j}, \qquad (3.22)$$

where
$$J_{g,j} = -\rho D_{m,j} \frac{\partial Y_{g,j}}{\partial x_i} - \frac{D_{T,j}}{T} \frac{\partial T}{\partial x_i}, \qquad (3.23)$$

Here, Sc_t is the turbulent Schmidt number, and its value is set to 0.85. *R* denotes the rate of production and destruction of gas species. D_m and D_T are the mass diffusion and the thermal diffusivity of gas species, respectively. To compute the diffusion fluxes, *J*, Maxwell-Stefan and Fick's law diffusion coefficients [158], including the thermal diffusion [139] for light species are adopted.

Surface species coverage:

$$\frac{d\theta_{j}}{dt} = \frac{s_{j}}{\Gamma} = 0 \ (j=1, 2..., N_{s}), \tag{3.24}$$

where θ is the surface species coverage, *s* is the surface species molar production rate and Γ is the surface site density. The transient term vanishes at steady state, and the net production rate becomes zero.

To model heat transfer in solid, the heat conduction equation is as follows:

$$\frac{\partial}{\partial x_i} \left(\lambda_s \frac{\partial T_s}{\partial x_i} \right) = 0, \qquad (3.25)$$

where λ_s and T_s are the solid conductivity and temperature, respectively.

3.2.2 Boundary conditions

As shown in Figure 3.1, the catalytic reactor is defined in a two-dimensional Cartesian frame. It brings in premixed reactants of hydrogen and air through the inlet, initiates the reaction on the catalytic surfaces, and then releases the resulting products through the outlet. The inflow conditions are kept uniform in terms of the flow velocity, temperature, and species mass fraction, with an inflow turbulence intensity of 5% (estimated based on the flow Reynolds number). At the outlet, all the variables experience a zero-gradient condition, except for the pressure, which is maintained at an atmospheric level. The gassolid boundary conditions at the interface of the catalytic reactor are as follows:

- The flow velocity components, *u*=0 and *v*=0, in the *x* and *y* directions, respectively, as the no-slip conditions.
- For the turbulence parameter, $\varepsilon = 2v(\partial\sqrt{k}/\partial y)$ is employed.
- The gas phase species boundary conditions at the gas-solid interface are specified by
 [ρD_m(Y_g/y)]_w= M_gṡ_g, where M_g and ṡ_g represent the molecular weight and catalytic
 molar production rate of gas species. Here, the mean catalytic reaction rates are
 evaluated at the mean wall temperature and the corresponding mean concentration
 of gas species, and they are basically treated using a "laminar-like" closure as
 discussed in referenced work [102].
- The thermal boundary conditions at the interface are set coupled.
- The outer walls, including the vertical facets of the plates, are considered adiabatic.
- The thermal conductivity of $\lambda_s = 16.27 \text{ Wm}^{-1}\text{K}^{-1}$ is used to account for heat conduction in solids.
- As the outer walls are adiabatic, the radiation of the hot catalytic surfaces towards the inlet and outlet is simulated using the discrete ordinates (DO) model. The DO model is uncoupled, and a value of 10 is set for the energy iterations per radiation iteration.
- The radiation heat transfer is considered at the boundary temperature at the inlet and outlet enclosures with an internal emissivity of 1.0.

Computations are performed on eight configurations (C1-C8), as shown in Figure 3.1 with the design parameters listed in Table 3.1. The L_i and L_b in reactor configuration denote the length of the inert zone and the catalytic zone, respectively. The configuration C1 is a planar reactor, which consists of two parallel plates with a thickness (δ_s) of 0.5 mm placed at a distance h, where the inner surfaces are Pt-coated. In the previous studies [76,109], it was found that the gap between the plates has a significant impact on the mass and heat transfer rates as well as on the catalytic process. To investigate this effect, we vary the value of h to 1-, 2-, 3-, 5-, and 7-mm. Again, a similar planar configuration C1 of h=3.5-mm is chosen to serve as a reference for comparisons with modifications considered for catalyst optimisation. Configuration C2 features half cylinders (HC) of diameter d=1.5-mm, symmetrically placed on the inner surfaces of the planar reactor with a distance dbetween them. The catalytic surfaces consist of 15 circular surfaces of HCs and 15 flat surfaces (P). C3 includes full cylinders (FC) of diameter d at the mid-plane of the reactor with a distance d between them, and the circular surfaces of 11 FCs and both the upper and lower inner surfaces of the planar reactor are defined as the catalytic surfaces. However, C4 is a combination of HC and FC placed alternately inside the reactor. The HCs are attached to the upper and lower inner surfaces and symmetrically placed. The FCs are placed in the mid-plane of the reactor. Here, the circular surfaces of 12 HCs and 12 FCs are the catalytic surfaces.

Catalytic	h	L_i	L_b	Scat	Re	Residence time
Reactor	(mm)	(mm)	(mm)	(mm^2)		(s)
C1	1	0	350	700	4200	0.010586
	2	0	500	1000	4200	0.030248
	3	0	700	1400	4200	0.063520
	5	0	900	1800	4200	0.136116
	7	0	1100	2200	4200	0.232909
	3.5	5.25	75	115	4353	0.006927
C2	3	5.25	75	115.69	3680	0.006927
C3	3	5.25	75	114.84	3811	0.006927
C4	3	5.25	75	113.1	3385	0.006927
C5	3	5.25	75	84.82	3385	0.006927
C6	3	5.25	75	70.69	3385	0.006927
C7	3	5.25	75	56.55	3385	0.006927
C8	3	5.25	75	28.27	3385	0.006927

Table 3.1 Computational conditions
However, the number of HCs and FCs for C2, C3, and C4 is considered to keep approximately the same amount of catalyst surface as C1. The C5, C6, C7, and C8 configurations are the same as C4 but with a reduction of 25%, 37.5%, 50%, and 75% catalyst from C4, respectively. The catalytic surface for these configurations is selected on specific locations of the circular surfaces where heat and mass transfer are effective.



Figure 3.1 Schematics of the various catalytic reactors. The red colour denotes the catalytic surface in the catalytic zone

A Finite Volume Method (FVM) is used to solve the governing equations for turbulent flow variables. Ansys Fluent 2023 R2 version was used for computations, with a 2-D steady-state planar and pressure-based solver. The LR variant of the k- ϵ model was set as the viscousmodel, and the species transport model was used to solve the volumetric and wall surface reactions. The chemistry solver was stiff, and the integration method was ISAT. The reaction rates were computed using Eddy-dissipation-concept (EDC). The SIMPLE scheme was used for pressure velocity coupling. For spatial discretisation, least square cell-based, and second-order method were set for gradients and pressure, respectively. A second-order upwind method was used for all other variables. The solution convergence criteria of 10⁻⁶ were set for all the residuals.

3.2.4 Chemical kinetics

To model the process of hydrogen catalytic combustion, we used both gas-phase and surface detailed reaction mechanisms. Our focus was on finding the appropriate gas-phase mechanism that could accurately predict the location of the flame, as the ignition process is complex and subject to debate. We investigated five different gas-phase mechanisms by Warnatz [141], Marinov [159], Li [160], kim [161] and Fureby [162]. For the surface mechanism, we employed the model developed by Deutschmann *et al.* [140], which simulates H₂ oxidation over Pt through three reversible and eleven irreversible reactions involving five surface and six gas-phase species. The gas-phase and surface reaction rates were evaluated using CHEMKIN [142] and Surface-CHEMKIN [143], respectively and transport properties were calculated using CHEMKIN transport database [144]. However, the mass-weighted mixing law was used to compute the mixture gas viscosity and thermal conductivity. For mass diffusivity and thermal diffusion coefficient, we utilised the kinetic theory. Overall, our approach enabled us to accurately model hydrogen catalytic combustion.

3.2.5 Mesh resolutions

The computational domain for C1 with varying *h* is considered comparatively long to ensure complete combustion. For h=7 mm, a structured grid of 1500×150 points (in the *x*- and *y*-directions, respectively) is used for the fluid domain, with refinement towards the inlet and walls. The solid domain is discretised with the 1500×30 grid points (in the *x*- and *y*-directions, respectively). A similar grid resolution is used for the other configurations as grid requirements for h < 7mm are less strict. For configurations C2, C3, and C4, a separate grid resolution test is conducted due to their distinct design conditions. All the configurations have several irregular surfaces with high gradients of flow variables, requiring grid

refinement near the walls and irregular surfaces. For comparison, three grid sizes of each configuration are taken and denoted as M1, M2, and M3 (Table 3.2).

For C2 in Figure 3.2 (a), the variation of catalytic temperatures along the reactor length and transverse mean H₂ distribution (at x=7.5 mm and 9 mm) are compared and it shows no significant changes among the tested grid sizes. Similarly, for C3 and C4, the grid test is performed using the centreline temperatures along the reactor length and transverse mean H₂ profiles (at x=7.5 mm and 9 mm) shown in Figure 3.2 (b) and Figure 3.2 (c), respectively. However, based on the comparisons shown and the precision requirement, the grid size M2 for the respective configuration is chosen for the computations.

Table 3.2 Number of grids tested

Туре	C2		C	3	C4	
	Fluid	Solid	Fluid	Solid	Fluid	Solid
M1	120305	62376	150460	79845	124307	85998
M2	176625	120874	198625	121878	175627	121878
M3	236705	148786	262020	144685	234147	162558



Figure 3.2 Effect of grid resolution for the configurations of C2, C3 and C4, (a) temperature on catalytic surface, T_{cat} (c, e) midplane temperature inside the reactor, T (b, d, f) transverse profile of H₂ mass fraction at location x=7.5 mm and 9 mm

The steady-state solutions obtained from the numerical calculations were validated by comparing them with the experimental results of Appel et al. [102]. The same reactor configuration was adopted as that used in the experiment. The operating parameters were replicated, and the measured wall temperatures were treated as the wall boundary conditions. The validated results presented in Figure 3.3 show the mean values of the species (H₂ and H₂O) and temperatures at the various axial locations. The results compare five gas-phase mechanisms with the experimental dataset. The objective here is to attain a suitable mechanism for the prediction of a realistic catalytic process. At x=25-mm, all the mechanisms accurately capture the X_{H2} and are in good agreement with the measurements. However, they underpredict the X_{H2} far from the walls near the reactor centre region with increasing x. Overall, the predictions of Warnatz [141], Kim [161], and Li [160] are similar, with Marinov [159] and Fureby [162] showing higher underprediction far downstream. Warnatz [141], Kim [161], and Li [160] produced similar predictions of H₂O and T, but they underpredict near the walls with increasing x. Compared to them, Marinov [159] and Fureby [162] have less underpredictions near the walls. To better justify the mechanisms, the OH contour plots in Figure 3.4 are presented along with the numerical and experimental results of Appel et al. [102]. The onset of gas-phase ignition (indicated by the vertical downward arrow) and OH ppmv level are taken into consideration for comparisons. The results indicate that an early ignition occurs with almost all the mechanisms including the numerical results of Appel et al. By comparison, the ignition locations of Warnatz [141] and Kim [161], are nearly similar and closer to measurement.

However, the underpredictions obtained are approximately 21.7%, 23.5%, 40.1%, 61.7%, and 66.8% for Warnatz [141], Kim [161], Li [160], Marinov [159] and Fureby [162], respectively. Similarly, the percentage variations of maximum OH ppmv among these mechanisms are 3.33%, 62.08%, 42.5%, 139.58%, and 107.08%, respectively. Overall, the Warnatz [141] mechanism provides reasonable agreement with the experimental measurements and is therefore chosen for the present study.



Figure 3.3 Comparisons of species and temperature mean profiles at different axial positions at φ =0.18, T_{in} =300 K, Re =15390, experiment (Appel et al [102])



Figure 3.4 OH contour (ppmv) (arrow indicates the onset of gas-phase ignition) at φ =0.18, *T_{in}*=300 K and *Re* =15390, exp (Appel et al [102])

3.4 Results and discussion

Firstly, we present the results of the combustion of fuel-lean premixed H_2/air in a catalytic planar reactor with varying *h* (i.e., the gap between the walls). The aim is to provide an indepth understanding of catalyst reduction and to facilitate the discussion of reactor development with an optimum use of catalysts under similar operating conditions. Next, we discuss the distribution of the Nusselt and Sherwood numbers on catalytic surfaces across different configurations, followed by a comparison of H_2 consumption rates.

3.4.1 Planar reactor configurations

Figure 3.5 shows the predicted H₂ conversion for each *h*. We define H₂ conversion as follows:

H₂ conversion (%) =
$$\frac{Y_{H2,avg,in} - Y_{H2,avg,x}}{Y_{H2,avg,in}} \times 100$$
 (3.26)

where $Y_{H2,avg,in}$ and $Y_{H2,avg,x}$ are the average H₂ mass fraction at the inlet and axial positions, respectively. In Figure 3.5 (a), the H₂ conversion reaches nearly 99.9% across the entire length of the catalytic reactor for various reactor heights (*h*). The results indicate that as the reactor height (*h*) decreases, a shorter catalytic reactor length is required to achieve complete H₂ conversion. This is due to the intensified diffusion of H₂ species with decreasing *h*, as discussed in Figure 3.5 (b). The figure illustrates the transverse distributions of H₂ and H₂O at *x*=10-mm for all the planar configurations. As *h* decreases, *Y*_{H2} decreases because of the strong diffusion of H₂ towards the reactor walls. This enhanced diffusion leads to greater H₂ consumption in the catalytic process, resulting in increased H₂O production at lower *h*.

The catalytic wall temperature (T_w) along the reactor length for different reactor heights (h) are shown in Figure 3.5 (a). The peak T_w values are located close to the upstream and shifts along the reactor length with increasing h. It is observed that a significant length of catalytic surface reaches superadiabatic temperatures, a common occurrence in H₂ catalytic combustion under nearly adiabatic conditions. Operating a reactor under such conditions is considered unfavourable, as discussed in literature [102,129]. The variation in T_w along the reactor length is a result of heat transfer from hot surface to incoming mixtures, as well as heat conduction in solid. The H₂ conversions in Figure 3.5 (a) for different reactor heights are obtained by keeping the same inflow conditions.





Figure 3.5 Planar catalytic reactor: (a) left axis: H₂ Conversion; right axis: temperature distribution on catalytic wall (T_w) (b) H₂ and H₂O distribution at *x*=10 mm, φ =0.15, T_{in} =300 K, *Re* =4200.

Planar Reactor (C1)	<i>h</i> =1 mm	<i>h</i> =2 mm	<i>h</i> =3 mm	<i>h</i> =5 mm	<i>h</i> =7 mm
\dot{m}_{H2} inlet (×10 ⁴ kg/s)	1.63921	1.63921	1.63921	1.63921	1.63921
Gas. conv. (× 10^4 kg/s)	0.00249	0.02899	0.10915	0.19817	0.23327
Cat. conv. (×10 ⁴ kg/s)	1.63672	1.61022	1.53006	1.44104	1.40594
Gas. conv. (%)	0.15	1.77	6.66	12.09	14.23
Cat. conv. (%)	99.85	98.23	93.34	87.91	85.77

Table 3.3 Gaseous and catalytic conversion rates at different planar reactor heights

However, the contributions of the gaseous reaction and the catalytic reaction pathways H_2 to conversion vary with the reactor heights (*h*), as shown in Table 3.3. For the reactor of h=1mm, the combustion mode is entirely catalytic. As *h* increases, the contribution of gaseous combustion also rises. In order to assess the effect of gaseous combustion at different reactor heights, some calculations are performed while maintaining constant operating conditions, but without gas-phase chemistry.

Figure 3.6 illustrates the changes in surface coverages of H(s), O(s), and OH(s) on the catalytic surface. For convenience, pure catalytic chemistry and coupled chemistry are referred as PC and CC, respectively. The surface coverage of H(s) and O(s) are controlled by the adsorption rate and near wall concentrations of H₂ and O₂, respectively. Again, under fuel lean condition, the O(s) is the main coverage, and is strongly dependent on wall temperature. At high temperatures, as shown in Figure 3.6 (b), the O(s) is low, which promotes H_2 adsorption. As temperature decreases, the O(s)increases while H(s) in Figure 3.6 (a) decreases due to lower concentrations of H₂. The effect of gas-phase chemistry becomes more significant as reactor height (*h*) increases. For h=1-mm, there is no variation between CC and PC, indicating purely catalytic combustion. At a higher reactor height of h=3-mm, a variation in H(s) is observed at peak values within a very limited catalytic length, along with changes in O(s) and OH(s). This suggests that gas-phase chemistry has a small contribution on the catalytic process at this height. However, for h = 5 mm and 7 mm, the effect of gas-phase chemistry becomes more substantial over a broader catalytic length. The flame inside the reactor is attached to the catalytic wall, which prevents the H₂ adsorption, resulting in lower H(s) in CC within the flame regime. Within the reactor, the flame is attached to the catalytic wall, which hinders H_2 adsorption, leading to lower H(s) in CC within the flame region. Conversely, excess O₂ promotes O₂ adsorption in the flame region, increasing O(s) coverage. Similarly, OH adsorption rises because the catalyst acts as a



sink for homogeneously produced OH, as discussed in earlier studies [150]. Beyond the flame region, the reaction shifts entirely to the catalytic mode.

Figure 3.6 Surface coverages (a) H(s) (b) O(s) (c) OH(s), $\varphi = 0.15$,

T_{in}=300 K, *Re* =4200

3.4.2 Conceptual design modifications for optimal catalyst

3.4.2.1 Combustion characteristics

Computations on the modified reactors are now being carried out using the same numerical approach as the planar reactor. Figure 3.7 presents the contours of the species mass fraction such as H₂, H₂O, and OH, as well as the flame temperatures for configurations C1, C2, C3, and C4. The computation maintains almost the same residence time and the same catalyst surface area for these reactors to establish a clear understanding of how each configuration affects the catalytic process. The planar reactor C1 of h=3.5-mm is used as a reference for comparisons.

Compared to C1, it is observed from the Y_{OH} contour in Figure 3.7 (c) for C2 that the gasphase chemistry plays a strong role in H₂ combustion, where the flames are ignited and stabilised near the inlet on the non-catalytic walls. The flames are also evident in the locations between the two half cylinders (HC). As a result, the Y_{H2} consumption and Y_{H2O} production in the flame regime is high, as also shown in Figure 3.7 (a) and Figure 3.7 (b), respectively. Additionally, the *T* contour in Figure 3.7 (d) indicates that the temperature increases in the solid domain due to a combination of gas-phase and catalytic combustion, which leaves the reactor core at a low temperature. However. The thickness of the solid plays a crucial role in catalytic reacting flows, particularly when subjected to temperature fluctuations, as highlighted in the work by Arani *et al.* [114]. As discussed in their article, a wall thickness of 50 microns allows the solid to closely track the temperature fluctuations imposed by the reacting flow. In the current modelling, however, we employ a significantly thicker wall of 0.5 mm (500 microns). This increased thickness enhances the solid's thermal inertia, thereby limiting the potential for fluctuations in the wall temperature.

Figure 3.7 (c) shows that the flames of C3 are ignited on the non-catalytic walls but are not as strong as C2. In the catalytic regime, no significant Y_{OH} is observed, indicating that the combustion mode is purely catalytic. Comparatively, the significant Y_{H2} consumption is observed in the flame regime, specifically on the catalytic flat surfaces between the two full cylinders (FC), as shown in Figure 3.7 (a). Consequently, the Y_{H2O} production is higher in those locations, as depicted in Figure 3.7 (b). In addition, the *T* contour in Figure 3.7 (d) shows that the catalytic process influences the temperature distributions in solid, thus high temperatures in the solid regime are observed in both the flat and cylinder domains up to x=13d, then gradually decrease with *x* as a low H₂ concentration for the catalytic process.



Figure 3.7 Contours of (a) Y_{H2} (b) Y_{H2O} (c) Y_{OH} and (d) T for reactors C1, C2, C3 and C4. Reactor length from x=0 to 57.5-mm is presented, φ =0.15, T_{in} =300 K.

When comparing the Y_{OH} of C4 to that of the other configurations in Figure 3.7 (c), the Y_{OH} contribution is insignificant over the domain. This means that no gas-phase ignition is observed in this case, and the mode of combustion is purely catalytic. Therefore, the Y_{H2} consumption takes place on the catalytic surfaces shown in Figure 3.7 (a), and the Y_{H2O} production in Figure 3.7 (b) is higher near the regime of catalytic surfaces. Regarding the temperature (T) in Figure 3.7 (d), it is almost uniform in the solid domain except in the first two full cylinders where the temperature is comparatively high. However, the orientation of HCs, FCs or both combinations inside the reactor has a significant effect on the flow field and influenced the heat and mass exchange between the flow and catalytic surfaces. Figure 3.8 displays the flow patterns along the length of the catalytic reactor. The reactor length is presented here only from x=0.25-mm to x=25.5-mm. For C2, the flow gets separated downstream of HC and vortices are formed. The vortices are located between two HCs and attached to both the upper and lower walls which are almost symmetrical about the mid plane. Similarly, the flow separation for C3 occurs downstream of FC, and the region between two FCs is covered by the vortices. For C3, the vortices are formed downstream of HCs and FCs, but the vortices size are comparative smaller than that of C2 and C3. Again, the branch of vortices near upstream of HCs is smaller in size than the vortices near downstream of HCs. Furthermore, there is no vortices observed at the upstream of FCs. After all, the vortices have significant effect on the heat transfer, and mass transfer of fuel, thus influencing the catalytic combustion process.



Figure 3.8 Streamlines for reactors C2, C3 and C4. Reactor length from x=2.5-mm to 25.5-mm is presented.

3.4.2.2 Heat and mass transfer

To assess the heat and mass exchange between the flow and catalytic surfaces, the Nusselt (Nu) and Sherwood number (Sh) are considered, and their definition are given as follows:

Nusselt Number:

$$Nu = \frac{h_c(2h)}{\lambda_g},$$
(3.27)

where h_c is the heat transfer coefficient and λ_g is the fluid conductivity. The expression for h_c is as follows:

$$h_{c} = \frac{\lambda_{g} \left. \frac{\partial T}{\partial y} \right|_{w}}{T_{w} - T_{ref}}, \qquad (3.28)$$

where $\partial T/\partial y$ is the gradient of temperature obtained normal to the surface. The T_w and T_{ref} are the surface and the reference temperatures, respectively. For convenience, T_{ref} is considered as an average of T_{in} and T_{adb} .

Sherwood Number:

$$Sh = \frac{k_c(2h)}{D_m},\tag{3.29}$$

where k_c and D_m are the H₂ mass transfer coefficient and mass diffusivity, respectively, and k_c is expressed as follows:

$$k_{c} = \frac{D_{m} \frac{\partial C_{H2}}{\partial y}\Big|_{w}}{C_{H2,w} - C_{H2,ref}} , \qquad (3.30)$$

where $\partial C_{H2}/\partial y$ is the gradient of H₂ concentration obtained normal to the surface. $C_{H2,s}$ and $C_{H2,ref}$ are the surface and the reference concentrations of H₂, respectively. $C_{H2,ref}$ is considered as an average of concentrations at T_{in} , and at T_{adb} where H₂ is completely consumed. Figure 3.9 shows the local Nu distribution on the catalytic surfaces for different configurations. Here, the Nu allows to assess the effectiveness of convective heat transfer processes from hot catalytic surfaces. For configuration C1, the Nu is initially high on the catalytic surfaces and gradually decreases along the length of the

reactor. However, for configuration C2, a significant variation of Nu is observed on the circular surfaces of the half cylinders (HC), and Nu is very small on the flat surfaces. There are some locations where Nu becomes negative, and this is because of the heat transfer from fluid to solid. Compared to C2, the Nu is higher on the surfaces of the full cylinders, and on the flat surfaces in configuration C3. For both cases (C2 and C3), Nu on the cylinder surfaces is low at very upstream and downstream of the cylinder and becomes at the maximum upper and lower regimes of the cylinder. Finally, for C4, Nu distributions on the cylinder surfaces are observed to be high and effective among the configurations compared.



Figure 3.9 Nusselt Number (*Nu*) on catalytic surfaces for configurations C1, C2, C3 and C4, P – planar, HC – half cylinder, FC – full cylinder

Figure 3.10 presents the local *Sh* values for the different configurations. Here, the *Sh* measures the effectiveness convective H_2 mass transfer towards the catalytic surfaces. In C1, the *Sh* value sharply decreases at the beginning of the catalytic surfaces due to the high concentration of H_2 clustered near the hot non-catalytic surface. Within a short catalytic length, the *Sh* value drops significantly and remains small with increasing *x*. Comparatively in C2, the variation of *Sh* is observed on circular surfaces, while the contribution on flat surfaces is minimal. This indicates that the catalytic

process is more effective on circular surfaces than on flat surfaces. Again, the negative Sh is the result of a negative gradient of H₂ downstream of the catalytic surfaces. Similarly, in C3, the *Sh* value is higher on circular surfaces except for the downstream portion, and the behaviour on flat surfaces is similar to that of C1. Overall, the *Sh* distributions on the catalytic surfaces of C4 are comparatively higher and effective for higher catalytic activity.



Figure 3.10 Sherwood Number (*Sh*) of H₂ on catalytic surfaces for configurations C1, C2, C3 and C4, P – planar, HC – half cylinder, FC – full cylinder

3.4.2.3 H₂ catalytic conversions

Figure 3.11 and Figure 3.12 show a comparison of the catalytic performance among the configurations listed in Table 3.1. The residence time and the amount of catalyst are again kept similar for configurations C1, C2, C3 and C4. The other configurations C5, C6, C7 and C8 are reactors with reduced catalysts, where the catalyst surface area from C4 is decreased for catalyst optimisation. The observations are listed as follows: Figure 3.11 (a) presents the H₂ conversion of reactors having the same catalytic surface area. By comparison, the C1 has the lowest performance over the reactor length and achieved a 62.6% conversion at the end of the catalytic region. After that, no conversion occurs because of the absence of the catalyst. For C2, however, the H₂ conversion plot is not as smooth as C1 and has peaks along the reactor length. The peaks and troughs of conversion are attained on the circular and flat surfaces, respectively. The H₂ conversion is effective over the circular surfaces as a result of an increased mass transfer, discussed in Figure 3.10. Due to the same reason, there is a high peak value of conversion for C3 in the region between the flat surfaces and cylinders. Comparatively, the mass transfer to the cylinder surfaces of C3 is more effective than C2, therefore, the H_2 conversion values are higher in C3. At the end of catalytic region, the H₂ conversion is achieved by 74.4% and 89.6% for C2 and C3, respectively. For C4, the H₂ conversion plot is smooth like C1, and this has the highest possible conversion of 93.3% at the end of catalytic region. Again, C4 has a lower conversion than C3 over the region of 5.12 mm < x < 21 mm though the mass and heat transfer over that reactor length is effective in C4 as depicted Figure 3.9 and Figure 3.10. This is because of the relative amount of catalyst coated along the reactor walls which is comparatively high for C3. However, the reactor shape significantly influences the contributions of gaseous and catalytic reaction pathways to H₂ conversion. As shown in Table 3.4, gaseous conversion in configuration C2 is almost double that of configuration C1, while it is minimal in configuration C3. In configuration C4, the combustion is purely catalytic. Figure 3.11 (b) shows the H₂ conversion of reactors with varying catalytic surface area. The configuration C4 is chosen for the catalyst optimisation analysis as this has a better heat and mass transfer capability. The additional four configurations C5, C6, C7 and C8 are the same as C4 but have the catalytic surface area of 75%, 62.5%, 50% and 25%, respectively. The orientation of catalytic surfaces is shown in Figure 3.1. However, the catalytic surfaces are selected in those locations where the H_2 mass transfer is relatively high.



Figure 3.11 H₂ Conversion: (a) almost same catalytic surface area, $S_{cat} \approx 0.15 \text{ m}^2$ (b) Varying S_{cat} .

As expected, the H₂ conversion plots of C5, C6, C7 and C8 are identical with C4 and the conversion values over the reactor length are comparatively low than the immediate reactor of high percentage of catalytic surface. But the catalyst reduction in each reactor configuration is very significant compared to the loss of H₂ conversion. At the end of catalytic region, the H₂ conversion is achieved by 91.08%, 89.04, 85.83% and 67.56% for C5, C6, C7 and C8, respectively. Compared to C4, with a 50% and 75% reduction in catalyst, this results in a 7.47% and 25.74% reduction in H₂ conversion, respectively. However, the maximum H₂ catalytic conversion rate per unit area is achieved with a 50% catalyst reduction in the C7 reactor reported in Table 3.4.

To assess the reactor performance with a reduced catalyst, Figure 3.12 shows the H₂ conversion at different percentages of the catalytic surface area compared to C1 (S_{cat} / $S_{cat,C1}$). It is noted that the H₂ conversion values are taken keeping the same operating condition as mentioned in Table 3.1, and estimated at the different axial locations of the reactor, therefore, the residence time at the axial locations is not the same in all the cases. However, the comparison for the catalyst surface area is still useful as the overall residence time in all the reactors are same. As shown in Figure 3.12, the H₂ conversions for C1 at different percentages of catalyst are lowest. Comparatively, for C2, there is an improved performance. However, the reactors C3-C8 have similar H₂ conversions which are significantly high compared to C1 and C2. At ~25% catalyst, the H₂ conversion is in the range of 67-71% for C3-C8. When the catalyst percentage is increased to ~50%, the H₂ conversion increases to 77% and 81-86% for C3 and C4-C7, respectively.

Reactor	C1	C2	C3	C4	C5	C6	C7	C8
\dot{m}_{H2} inlet	1.698	1.436	1.487	1.321	1.321	1.321	1.321	1.321
$(\times 10^4 \text{kg/s})$								
\dot{m}_{H2} outlet								
$(\times 10^4 \text{kg/s})$	0.635	0.367	0.154	0.088	0.117	0.145	0.187	0.428
Gas. conv.	0.234	0.379	0.051	1E-5				
$(\times 10^4 \text{kg/s})$					4E-7	9E-6	2E-5	4E-5
Cat. conv.	0.829	0.689	1.280	1.232				
$(\times 10^4 \text{kg/s})$					1.203	1.175	1.134	0.892
Cat. conv. per								
unit area (kg/s)	0.720	0.595	1.115	1.090	1.418	1.663	2.005	3.157
Gas. conv. (%)	13.79	26.41	3.49	0.00	0.00	0.00	0.00	0.00
Cat. conv. (%)	48.80	47.97	86.09	93.31	91.09	89.04	85.84	67.56

Table 3.4 Gaseous and catalytic conversion rates at different catalytic reactor



Figure 3.12 H₂ conversion as a function of catalyst amount $(S_{cat}/S_{cat,Cl})$

Again, a further increment to ~75% catalyst results in H₂ conversion of 86-91% for C3-C6. For ~100% catalyst, the maximum conversion is 89-93% for C3-C4. To summarise, the reactors with combined HCs and FCs are proved to be effective for an improved catalytic conversion. With segmented coating on HCs and FCs at a similar amount of catalytic surface, the change in H₂ conversion is within 4-5%. However, this key finding is crucial for developing a cost-effective catalytic reactor. Finally, the catalytic reactors investigated have significant variation in the pressure drop, as listed in Table 3.5. The

pressure drop here is characterised by the percentage decrease from that of the inlet pressure. The high-pressure drop is a consequence of flow separation and constricted flow. However, in catalytic reactor design, finding an optimal balance between H_2 conversion efficiency and pressure drop is essential, as these factors directly influence reactor throughput, energy efficiency, and operational costs. To achieve this balance, future work could focus on optimising design aspects such as reactor geometry, including cylinder size and positioning, improving flow distribution, and selecting effective catalyst coating strategies.

Catalytic	Flow	Reynolds	Dimensionless		
Reactor	Volume	Number	Pressure Drop (%)		
	$(10^{-6} \times m^3)$				
C1	262.5	4353	31.59		
C2	198.49	3680	79.79		
C3	205.56	3811	90.26		
C4	182.58	3385	98.43		
C5	182.58	3385	98.43		
C6	182.58	3385	98.40		
C7	182.58	3385	98.39		
C8	182.58	3385	98.30		

Table 3.5 Pressure drops

3.5 Conclusions

Premixed turbulent combustion of highly lean and ultra-lean hydrogen/air mixtures in a planar/non-planar reactor with platinum coating on inner surfaces was numerically investigated. Computations were carried out in multicomponent species transport model of turbulent solver using Ansys Fluent. Prior to analysis, the model was validated with available experimental results. To intensify the catalytic process, the catalytic surfaces are modified in such way to have increased mass and heat convection. The key findings of this work are:

- The reactor with high S/V ratio has significant impact on catalytic process and could burn H₂ completely within shorter length of the catalytically coated region.
- For non-planar configurations, the combined use of half and full cylinders was shown to significantly enhance mass and heat convection, resulting in an overall superior catalytic performance.

- The surfaces where high mass and heat transfer occurred can be coated optimally. A modified non-planar reactor can increase H₂ conversion by 30.7% compared to an equivalent planar reactor.
- Discrete coating in a non-planar reactor has been proven as a useful approach for catalyst coating optimisation to save catalyst by 50% while achieving a catalytic conversion rate of 2 kg/s per unit surface area of the catalytically-coated region.

However, the turbulence near the catalytic surfaces and the flow separation or wake formed in non-planar reactors significantly affect the catalytic process. Since the RANS equations have limitations in predicting the realistic turbulent characteristics in catalytic processes, the LES is used to analyse these characteristics in the next chapter. For this analysis, the first half portion of the catalytic region of configuration C5 is selected as it demonstrates higher catalytic conversion compared to other configurations with discrete coatings.

Chapter 4: A platinum-coated staggered reactor to intensify lean hydrogen/air combustion: a large eddy simulation study

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Abstract

Catalytic-aided combustion has been proven effective for premixed hydrogen/air mixtures, particularly under lean to ultra-lean conditions. However, minimising the required catalyst sets a significant challenge because noble metals with high catalytic activity are rare and expensive. Therefore, this study aims to intensify the catalytic combustion process by investigating a non-planar reactor comprising an array of platinum-coated half- and fullcylinders through large eddy simulation. A premixed mixture with a fuel-lean equivalence ratio of 0.15 and an incoming Reynolds number of 3500 based on hydraulic diameter is used. For comparison, a planar reactor without cylinders is also studied under the same operating conditions and with the same amount of platinum-coated surface area. The simulation employs the turbulent kinetic energy sub-grid model and the eddy dissipation concept to model the turbulent catalytic reacting flow. The discrete ordinate model is used to account for radiation heat transfer in the catalytic process. Numerical simulations are validated against experimental results prior to analysis. The findings indicate that the placement of cylinders along the reactor length enhances convective mass transfer and intensifies catalytic combustion, resulting in effective combustion over a smaller catalytic surface. Compared to planar models, non-planar reactors demonstrate a much better H₂ conversion efficiency throughout the reactor length, saving nearly 62.5% of the catalyst.

4.1 Introduction

Catalytic combustion of hydrogen is a modern technology and is being implemented in various reactor applications [50,70,119]. Among these applications, catalytic combustion of premixed mixtures has been found to be effective in reducing NO_x emissions [70,129] and in enhancing syngas combustion[63,163]. In previous studies [66,87,117,164], efforts have

been made to develop planar reactors with catalyst-coated inner walls. The fuel conversion inside the reactor is governed by either homogeneous combustion or heterogeneous combustion, or both based on inflow and reactor wall thermal condition [150]. The coexistence of homogeneous and heterogeneous combustion is advantageous due to reduced reliance on catalysts [165]. However, in reactors operated with lean mixtures, a significant amount of catalyst is required, with fuel conversion predominantly occurring through heterogeneous combustion [150]. This can result in superadiabatic surface temperatures at the catalytic walls [68,76]. In many cases, localised hotspots (where the local wall temperature exceeds the average temperature) can form if the support material is unable to dissipate heat effectively [53,92]. Although some recent studies [44,91,166] have explored catalyst and reactor designs aimed at minimising elevated wall temperatures and enhancing combustion stability, further research is needed to evaluate their effectiveness under diverse operating conditions.

Reducing the catalyst requirement for complete combustion presents a significant challenge due to the high cost of rare metals used as catalysts. Additionally, accommodating sufficient catalyst in planar reactors necessitates increased reactor length, leading to a more robust but less economical and practical design [150]. Moreover, in planar reactors, surface reaction rates often reach a plateau due to limited mass transfer of fuel species toward the catalytic walls [69,76]. Therefore, catalytic reactors with the enhanced mass transfer of fuel species to reactive walls, compact designs and optimal catalysts are of prime interest for their wide-ranging application [68,107]. To progress on catalytic reactors, the multidimensional modelling of optimal catalytic coating is necessary for reactor design.

One application of catalytic reactors is honeycomb structures, which consist of multitube catalyst-coated channels with a hydraulic diameter in the millimetre range. To enhance mass transfer with an optimal catalyst in honeycomb channels, a novel reactor design has been adopted with appropriate geometric modifications[167]. Carrying out experimental work in such reactors is exceedingly challenging, particularly in the precise measurement of species concentration and temperature near the catalytic wall [102,168]. Consequently, numerical simulation is the preferred approach for investigating catalytic processes. In this context, the proper selection of turbulence models is essential for accurately simulating the complex turbulent flows inside the reactor, because the turbulent effect becomes weaker when it involves the catalytic reaction with the flows [102]. Moreover, the turbulent flow significantly affects the catalytic activity as the mass transport toward or away from the catalytic surface increases with turbulence [102,116]. Therefore, a reliable numerical model is essential to capture the near-wall turbulence and the catalytic process. Reynolds Averaging

Navier Stokes (RANS) models are widely used [101,102,111] for modelling turbulent catalytic reacting flows. However, RANS models struggle to capture the effects of separated flow in reacting flows with non-planar surfaces. In this regard, Direct Numerical Simulations (DNS) are the best numerical models to capture near-wall turbulence, but this is computationally expensive and difficult to model complex combustion dynamics with catalytic reactions. Though some studies using DNS [113–115] for turbulent catalytic combustion systems have been reported in the literature, the reacting DNS is challenging for spatially developing flows, particularly for the reactor configuration discussed in the present study. The Large Eddy Simulations (LES) are, therefore, practical options to simulate most of the turbulent reacting flows [169]. Though the LES is limited to modelling large-scale turbulence, this is still useful for predicting realistic combustion processes [60,170,171]. Nonetheless, currently, there is a noticeable research gap in the application of LES to catalytic combustion processes. This work aims to address that gap by evaluating the effectiveness of LES in modelling catalytic combustion systems, highlighting its potential to enhance our understanding and prediction of these complex processes.

Previous studies [102,113–115] investigated the catalytic turbulent reacting flow in a channel or planar reactor, with an inflow Reynolds number of up to 30000. The high Reynolds number was considered to understand the effect of flow laminarisation, which occurs within the boundary layer and near the catalytic surface due to increased viscosity from heating by the catalytic surfaces. As reported in earlier studies [102], the laminarisation effect is expected to be more pronounced at low Reynolds number turbulent flows. However, the impact of flow laminarisation on catalytic reactors with non-planar surfaces is not yet known and requires further investigation. Therefore, comparatively a low turbulent Reynolds number of 3500 (based on the hydraulic diameter and the mean inlet velocity) is chosen to simulate the turbulent catalytic combustion with detailed chemistry (both heterogeneous and homogeneous). Simulations are performed on both planar and modified non-planar catalytic reactors at fuel lean H₂/air mixtures (equivalence ratios $\varphi = 0.15$).

The present work aims to intensify catalytic hydrogen/air combustion while reducing the reliance on expensive platinum catalysts. The innovation lies in the use of catalyst-coated staggered cylinders within the reactor to significantly improve the catalytic process. Furthermore, numerical simulations of turbulent catalytic combustion using LES are still limited, especially for reactors with cylindrical geometries as examined in this study, due to the challenges in accurately capturing the impact of reactor geometry on combustion dynamics. This study introduces a crucial design modification to catalytic reactors, paving the way for further advancements in catalytic combustion.

This article is organised in the following way: Section 4.2 explains the numerical methodology and presents validated results. Then, in Section 4.3, the discussion of the turbulent flow characteristics for both planar and non-planar configurations is presented, comparing the velocity, temperature and species distribution inside the reactor. Finally, the article concludes with a summary of the findings in Section 4.4.

4.2 Numerical approach

The catalytic reactor configuration, governing equations and numerical settings required to simulate turbulent catalytic reacting flow are discussed below.

4.2.1 Catalytic reactor configurations

The study investigates turbulent catalytic reacting flow in two reactor configurations. The first configuration is a planar one similar to the flow between parallel plates where a portion of inner surfaces is catalyst coated. It is noted that most previous studies on turbulent catalytic combustion have been focused on this configuration [114,115]. The second configuration is the non-planar reactor that represents the flow through an array of half- and full- cylindrical rods where a portion of cylindrical surfaces are coated with catalysts.



Figure 4.1 Schematics of the (a) planar and (b) non-planar reactors.

Such configuration has not been subjected to any study in the past. The computational domain is illustrated in Figure 4.1. The catalytic reactors are 11.67*H* in length and 1.5*H* in width, where *H* is the distance between the parallel plates and δ is the thickness of the plate. The cylinder pitch is denoted as *P* and its value of 2*d* is considered in the present case. The catalytic surfaces are highlighted in red in both configurations. The inner surfaces of 1.75 $\leq x/H \leq 8.82$ are coated with platinum in the planar reactor.

Case	C1. Planar	C2. Non-	C3. Planar		C4. Non-Planar	
(Reacting)		Planar (Non-	(Non-/Reacting)		(Reacting)	
		Reacting)				
Dimension	35.7 <i>H</i> × <i>H</i> ×1.5	10.5 <i>H</i> × <i>H</i> ×1.5	11 <i>H×H×</i> 1.5 <i>H</i> ;		11 <i>H</i> × <i>H</i> ×1.5 <i>H</i> ; <i>P</i> = <i>H</i> ;	
Н;		H;	<i>H</i> =3 mm;		<i>D=H</i> /2; <i>H</i> =3 mm;	
	<i>H</i> =7 mm	<i>P</i> =1.73 <i>H</i> ;	<i>δ</i> =0.5 mm		$\delta=0.5 \text{ mm}$	
		D=0.59H;				
		<i>H</i> =85.3 mm				
Grid	C1	C2	C3-M1	C3-M2	C4-M1	C4-M2
Fluid	3136000	7224000	756000	1225000	1477400	3826900
domain						
Solid	-	-	378000	525000	1246520	3520860
domain						
Inflow	<i>T</i> _{in} =300 K,		$T_{in}=300$	K, <i>φ</i> =0.15,	$T_{in}=3$	00 K,
conditions	<i>φ</i> =0.18,	<i>Re</i> =21304	Re=	3500	$\varphi=0$.15,
<i>Re</i> =15390					<i>Re</i> =3500	

Table 4.1 Simulated cases

The non-planar reactor contains six half cylinders attached to each reactor wall, and six full cylinders placed between the half cylinders along the centreline of the reactor. The centre of the first full cylinder is placed at a distance of 4*d* from the inlet. The distance between the centres of the half and full cylinders is *d*. The key design parameters for a non-planar reactor include cylinder size, aspect ratio, and spacing between the cylinders, all of which can significantly influence flow dynamics and catalytic performance. While the impact of variations in these parameters is not explored in the current study, the catalytic surfaces in the non-planar reactor are chosen where the mass and heat transfers are more effective. For this reason, only cylindrical surfaces are coated with platinum, except a quarter of the back uncoated. This is because flow separation occurs downstream of both half and full cylinders [167,172] which significantly affects the convective mass and heat transfer in that particular region. The total catalytic surface area for both configurations is kept the same for

comparison. Table 4.1 provides the other necessary information of geometric and operational parameters. The catalytic reactor operates by introducing a mixture of hydrogen and air into the inlet. The reaction is initiated on the catalytic surfaces and the resulting products are then released through the outlet. A Reynolds number (Re) of 3500 is used in both the catalytic reactors, based on the bulk velocity at the inlet and the hydraulic diameter. The inflow conditions such as flow velocity, temperature, and species mass fraction are kept uniform with a turbulence intensity of 5.7% estimated based on the flow Reynolds number. At the outlet, all the variables have a zero-gradient condition except for the pressure which remains at an atmospheric level. The gas-solid boundary conditions at the interface of the reactor are as follows:

- The flow velocity components, U=0, V=0, and W=0 as the no-slip conditions.
- The gas phase species boundary conditions at the catalytic surface are specified by $n_i[\rho Y_{g,j} (V_{g,j}+u_i)]_w = M_{g,j}\dot{s}_{g,j}, j=1, 2, 3,..., N_g$, where V_g , M_g , and \dot{s}_g are the diffusion velocity, molecular weight and catalytic molar production rate of gas species, respectively. n_i is the unit outward-pointing component normal to a surface, and u_i is the Stefan velocity. This Stephan velocity, arising from the net mass flux between the surface and the gas in catalytic process, is significant in transient formulations [173] and given by $n_i u_i = (1/\rho) \sum_{j=1}^{N_g} M_{g,j} \dot{s}_{g,j}$ [143]. However, for statistically steady-state solutions, the temporal fluctuations around the mean values do not generate significant Stefan velocities. Consequently, although u is nonzero, it is negligible in the present case.
- The thermal boundary conditions at the fluid and solid interface are set coupled.
- The outer walls and the vertical facets of the plates are considered to be adiabatic.
- The spanwise z-direction is subject to be a periodic boundary condition.
- The thermal conductivity of steel ($\lambda_s = 16.27 \text{ Wm}^{-1}\text{K}^{-1}$) is considered for heat conduction in solids.
- The process of thermal radiation from the hot catalytic surfaces towards the inlet and outlet is simulated using the discrete ordinates (DO) model. This model is uncoupled, and 10 iterations are set for energy per radiation iteration.
- The radiation heat transfer is considered at the inlet and outlet enclosures with an internal emissivity of 1.0 at the boundary temperature.

4.2.2 Governing equations

For LES computations, we use the filtered continuity, momentum, species, and energy equations as stated below. In all the equations, the filtered variable is denoted by an overbar.

$$\frac{\partial \rho}{\partial t} + \frac{\partial}{\partial x_i} \left(\rho \overline{u_i} \right) = 0, \qquad (4.31)$$

Momentum:

Continuity:

$$\frac{\partial}{\partial t} \left(\rho \overline{u}_i \right) + \frac{\partial}{\partial x_j} \left(\rho \overline{u}_i \overline{u}_j \right) = -\frac{\partial p}{\partial x_i} + \frac{\partial \sigma_{ij}}{\partial x_j} - \frac{\partial \rho \tau_{ij}}{\partial x_j}, \qquad (4.32)$$

where σ_{ij} and τ_{ij} are the stress tensors due to the molecular viscosity and the subgrid-scale (SGS) stress, respectively. Their definitions are given as follows:

$$\sigma_{ij} \equiv \mu \left(\frac{\partial u_i}{\partial x_j} + \frac{\partial u_j}{\partial x_i} - \frac{2}{3} \delta_{ij} \frac{\partial u_l}{\partial x_l} \right), \tag{4.33}$$

$$\tau_{ij} \equiv \overline{u_i u_j} - \overline{u}_i \overline{u}_j, \qquad (4.34)$$

For the sub-grid stress (SGS), the dynamic kinetic energy (DKE) model [174] is adopted, where the SGS kinetic energy is defined as $k_{sgs} = \frac{1}{2} \left(\overline{u_k^2} - \overline{u_k}^2 \right)$. The k_{sgs} is used to compute the SGS eddy viscosity, v_t defined as $v_t = C_k k_{sgs}^{1/2} \Delta_f$. Here, Δ_f is the filter-size calculated from $\Delta_f \equiv V^{1/3}$, where V is the volume of a computational cell. The SGS stress can then be rewritten in the following way:

$$\tau_{ij} - \frac{2}{3}k_{sgs}\delta_{ij} = -2C_k k_{sgs}^{1/2} \Delta_f \overline{S}_{ij}, \qquad (4.35)$$

where \overline{S}_{ij} is the rate-of-strain tensor for the resolved scale defined by

$$\overline{S}_{ij} \equiv \frac{1}{2} \left(\frac{\partial \overline{u}_i}{\partial x_j} + \frac{\partial \overline{u}_j}{\partial x_i} \right), \tag{4.36}$$

For the SGS kinetic energy, k_{sgs} the following transport equation is solved.

$$\frac{\partial \overline{k}_{sgs}}{\partial t} + \frac{\partial \overline{u}_{j} \overline{k}_{sgs}}{\partial x_{j}} = -\tau_{ij} \frac{\partial \overline{u}_{j}}{\partial x_{j}} - C_{\varepsilon} \frac{k_{sgs}^{3/2}}{\Delta_{f}} + \frac{\partial}{\partial x_{j}} \left(\frac{\nu_{t}}{\sigma_{k}} \frac{\partial k_{sgs}}{\partial x_{j}} \right), \tag{4.37}$$

The model constants in the above equations, C_k and C_{ε} , are determined dynamically, and σ_k is equal to 1.0.

Species transport:

$$\frac{\partial \rho \overline{Y}_{g,k}}{\partial t} + \frac{\partial}{\partial x_j} \left(\rho \overline{u}_j \overline{Y}_{g,k} \right) = -\frac{\partial J_{g,k}}{\partial x_j} + \overline{w}_{g,k} - \dot{w}_{sgs,k} - \frac{\partial \rho \gamma_{jsgs,k}}{\partial x_j} , \quad (k=1, 2, ..., N_g) , \quad (4.38)$$

where the species diffusion flux and the SGS mass flux are defined as follows:

$$J_{g,k} = -\rho D_{m,k} \frac{\partial Y_{g,k}}{\partial x_j} - \frac{D_{T,k}}{T} \frac{\partial T}{\partial x_j}, \qquad (4.39)$$

$$\gamma_{jsgs,k} = \overline{u_j Y_{g,k}} - \overline{u_j} \overline{Y_{g,k}} = \frac{\nu_t}{\sigma_{Y,k}} \frac{\partial Y_{g,k}}{\partial x_j} , \qquad (4.40)$$

Energy:

$$\frac{\partial \rho \bar{h}}{\partial t} + \frac{\partial}{\partial x_j} \left(\rho \bar{h} \bar{u}_j \right) = \frac{\partial}{\partial x_j} \left(\frac{\mu}{\Pr} \frac{\partial \bar{h}}{\partial x_j} - \sum_{k=1}^n \bar{h}_{g,k} \bar{J}_{g,k} \right) + S_h - \frac{\partial \rho q_{jsgs}}{\partial x_j}, \qquad (4.41)$$

where S_h is sources of energy due to chemical reaction. The diffusion fluxes, J_g are computed using Maxwell-Stefan and Fick's law diffusion coefficients [158] considering the thermal diffusion [139] for light species. The SGS heat flux is modelled as follows:

$$q_{jsgs} = \overline{u_j h} - \overline{u_j h} = \frac{v_t}{\sigma_t} \frac{\partial T}{\partial x_j} , \qquad (4.42)$$

In equations (9) and (11), the model constants σ_t and σ_Y are the SGS turbulent Prandtl number and Schmidt number, respectively which are obtained by the dynamic procedure proposed by Germano *et al.* [175].

Surface species coverage:
$$\frac{d\theta_j}{dt} = \frac{\dot{s}_j}{\Gamma} = 0$$
, $(j=1, 2..., N_s)$,
(4.43)

where θ is the surface species coverage, \dot{s} is the surface species molar production rate and Γ is the surface site density.

To model heat transfer in solid, the heat conduction equation is stated as follows:

$$\frac{\partial \rho_s h_s}{\partial t} + \frac{\partial}{\partial x_j} \left(\lambda_s \frac{\partial T_s}{\partial x_j} \right) = 0,$$

(4.44)

4.2.3 Solutions algorithm and solver setting

A pressure-based solver was employed in Ansys Fluent 2023 R2 to solve the governing equations for turbulent flow variables using the Finite Volume Method (FVM). The incompressible filtered Navier-Stokes and species transport equations were discretised on a structured hexahedral grid. The discretised equations were solved in both space and time using the SIMPLE algorithm. In LES setting, the dynamic kinetic energy (DKE) subgridscale model is used to model the sub grid scale turbulence developed by Kim and Menon [174]. This model solves the transport equation of subgrid kinetic energy where model constants are determined dynamically. For the transient formulation, the bounded secondorder implicit scheme was selected. For spatial discretisation, the least square cell-based method was used for gradients while the second-order method was used for pressure. The bounded central differencing scheme was used to compute convective fluxes. The stiff chemistry solver was chosen for thermochemistry, considering the integration method of ISAT (In situ adaptive tabulation). The Eddy-dissipation-concept (EDC) model of Gran et al. [176] was adopted to compute the gaseous reaction rates. Like EDC, a closure model is essential for accurately describing turbulent catalytic rates. Arani et al. [114,115] showed that turbulence significantly impacts the transport of species like H_2 and O_2 at the catalytic surface, especially at high Reynolds numbers of 11,400 and 24,600, which are notably higher than those in our study. They also found that solids with high thermal inertia do not respond to temperature fluctuations induced by the reacting flow. In our study, the substantial wall thickness ($\delta = 0.5$ mm) results in high thermal inertia, which effectively dampens these temperature fluctuations at the catalytic surface. As a result, variations in temperature and concentration at the catalytic surfaces are minimal compared to their mean values. This diminishes the significant reaction nonlinearity typically associated with the Arrhenius exponential term. When nonlinearity does occur, it remains minimal under constant wall temperature conditions [102]. Consequently, the catalytic rates are modelled using a "laminar-like" closure, evaluated based on the surface temperature and the corresponding gas concentrations, as outlined in the referenced work [102]. Detailed chemical reaction mechanisms of catalytic (heterogeneous) and gas-phase (homogeneous) were employed. For homogeneous chemistry, the reaction mechanism of Warnatz et al. [141] was used, while the heterogeneous was obtained from Deutschmann et al. [140]. The validation of coupled (hetero-/homogeneous) reaction mechanisms was reported in previous studies [102,150]. CHEMKIN [177] and Surface-CHEMKIN [143] were used to evaluate the gaseous and surface reaction rates, respectively, while the transport properties were calculated using the

CHEMKIN transport database [144]. However, we used the mass-weighted mixing law to calculate the viscosity and thermal conductivity of the mixture gas. To determine the mass diffusivity and thermal diffusion coefficient, the kinetic theory was applied.

Computations were performed on a High-Performance Computer (HPC) with Intel Xeon Gold 6138 (Skylake) processors at the ARCHIE-WeSt supercomputer centre using 40 cores. Computations were continued with a time step of 10⁻⁶s until the flow reached a statistically steady state. The integration time to reach the steady states was 8 and 12 flow-through times for the planar and non-planar reactors, respectively. The LES statistics were averaged around 6 flow-through times for all the results reported here.

4.2.4 Mesh resolutions

Both the planar and non-planar configurations use a hexahedral grid to discretise the domains as shown in Figure 4.2. In LES, large turbulent structures are directly resolved by the grid, while small-scale turbulence near the wall is captured using a subgrid-scale (SGS) model. Consequently, accurate wall treatment is essential in the LES methodology employed here to effectively capture the turbulent flow behaviour near solid boundaries. The grid resolution in the *y*-direction is mostly demanding and required to capture the most of flow fluctuations. In all cases, the near-wall grids are refined, with the first grid point placed at a nondimensional wall distance, y+ (where $y+=u_t\Delta y/v$) of less than 1.0. This allows the simulation to resolve the near-wall turbulence directly, without relying on wall functions. The spanwise z-direction has 70 grid points that are uniformly allocated, while the allocation of grid points in the streamwise *x*-direction is non-uniform. We used the grid resolutions (*x*-direction × *y*direction) of 640×70 , 180×60 , and 250×70 in fluid domain for case 1 (C1),



Figure 4.2 Example of grid generation.

case 3 (C3-M1) and case 3 (C3-M2), respectively, in the planar reactor. The first grid point from the walls was placed at y+=0.54 for case 1 and y+=0.45 for case 3 (C3-M1), respectively, based on the maximum y+. For case 2 (C2) and case 4 (C4-M1), the total grid points in the x-direction were 740 and 630, respectively. The grid points between the reactor flat wall and full cylinder along the y-direction were 55 for case 2 and 35 for case 4, while the points between the two half cylinders along the y-direction were 60 for case 2 and 50 for case 4. The nearest grid point at the cylinder was placed at y+=0.96 for case 2 and y+=0.92 for case 4.



Figure 4.3 Effect of grid resolution for the catalytic planar configuration (Case 3). Time averaged transverse profiles of (a) U_m and k, (b) T_m and T_{rms} , and (c) Y_{H2} and $Y_{H2,rms}$ at x/H = 7.5, $T_{in} = 300$ K, $\varphi = 0.15$ and Re = 3500.



Figure 4.4 Effect of grid resolution for the catalytic non-planar configuration (Case 4). Time averaged transverse profiles of (a) U_m and k, (b) T_m and T_{rms} , and (c) Y_{H2} and $Y_{H2,rms}$, at x/H=7.23, $T_{in}=300$ K, $\varphi=0.15$ and Re=3500.

Both C3 and C4 use solid domains to account for the conjugate heat transfer effect on catalytic combustion process. The number of grid cells for the simulated cases is summarised in Table 4.1. Figure 4.3 compares the mean profiles of axial *x*-component velocity, temperature, mass fraction of H₂ species, and their fluctuations at x/H=7.5 for two grids (C3-M1 and C3-M2) in the planar configuration. The comparison shows no significant difference

in U_m , T_m and $Y_{m,H2}$ between the two grids. However, the grid C3-M1 underpredicts fluctuations at the location of peak values. Similarly, for the non-planar configuration shown in Figure 4.4, a grid test is conducted at x/H=7.23 for two grid sizes (C4-M1 and C4-M2). In this case, the grid C4-M1 overpredicts U_m , T_m and $Y_{m,H2}$ at location of peak values as well as their fluctuations.

4.3 Validation of the numerical Solutions

The computational procedure of LES modelling in the planar and non-planar reactors shown in Figure 4.1 is initially verified by reproducing the experimental catalytic reactor (case C1) of Appel *et al.* [102] and non-catalytic reactor (case C2) of Smith *et al.* [167]. Then, the effect of catalytic turbulent reacting flow for both the reactors (case C3 and case C4) is discussed and compared considering the same inflow condition and catalytic surface area. All the statistical parameters presented have been averaged over time and spanwise *z*direction.

Figure 4.5 shows the transverse mean profiles of velocity, species (H₂ and H₂O), temperature, and turbulent kinetic energy at different streamwise locations for the catalytic planar reactor. The results are compared with the experimental data obtained from Appel *et al.* [102]. The present LES model captures well the continuous change of mean velocity (U_{avg}) and turbulent kinetic energy (k_{avg}) with increasing *x* in Figure 4.5 (a). The prediction of H₂ and H₂O over the reactor shown in Figure 4.5 (b) is in accord with the measurement data. However, there is a slight underprediction of H₂O at the far downstream near the wall regions. The reason is the limitation of the kinetic mechanism to produce the catalytic reaction process discussed in the previous studies [129]. Therefore, the underprediction of mean temperatures is observed at the similar locations shown in Figure 4.5 (c). However, the measurement uncertainty reported for the species and temperature is ±10% and ±50K, respectively [102]. Overall, the LES predictions are satisfactory, and the same LES model is used to simulate the catalytic turbulent combustion process.

The catalytic reacting flow through a non-planar reactor, shown in Figure 4.1 (b), has not been studied before. Therefore, for LES validation in the non-planar reactor, the experimental study [167] of non-reacting flow (case C2) is simulated and compared. Figure 4.6 shows the comparison of LES predictions for the mean velocity and turbulent quantities. The results are presented at the location of x/H=6.51, midway between the full and half cylinders as the experimental results reported only that location. The predicted *x*- and *y*- components of mean velocity shown in Figure 4.6 (a) and Figure 4.6 (b), respectively, are in

close agreement with the measurements. Again, the turbulent fluctuations in Figure 4.6 (c)-(e) are generally captured well with the LES model. The only exception is the peak values of V_{rms} at the reactor core, where the level of deviation from the experimental results appears to be considerable due to the flow separation and reattachment in the wake region.



Figure 4.5 Case C1 (reacting): LES comparison at various streamwise locations for the timeaveraged mean profiles of (a) U_m and k, (b) X_{H2} and X_{H2O} , and (c) T_m , at T_{in} =300 K, φ =0.18, Re=15390, experiment (Appel et al [102])




Figure 4.6 Case C2 (non-reacting): LES comparison for the time-averaged mean velocity (U_m and V_m) and Reynold stresses (R_{UV}) at x/H=6.51, Re=21304, experiment ([167])

4.4 Results and discussion

In this Section, the validated LES model is used for the investigation of catalytic reacting flow in both the planar (case C3) and non-planar reactors (case C4) keeping the same inflow Reynolds number of 3500, inflow temperature of 300 K, and an equivalence ratio of 0.15. The catalytic surface area of 42.41 mm² is kept the same for both the configurations to allow for a comparison and better understanding of the catalytic process.

To visualise the flow field, Figure 4.7 (a)-(c) illustrates the contours of the instantaneous axial velocity (U), mean axial velocity (U_m), and its fluctuations (U_{rms}) for the planar catalytic



Figure 4.7 Case 3 (reacting): Contours of (a) instantaneous x-velocity, U, (b) time-averaged mean x-velocity, U_m and (c) its fluctuations, U_{rms} onto the xy-plane at z/H = 0.75. Transverse profiles of time-averaged (d) U_m (e) k (f) U_{rms} (h) W_{rms} and (i) R_{UV} (or $U_{rms}V_{rms}$) at five streamwise locations.

reactor. In Figure 4.7 (b), it is shown that the mean velocity increases along the length of the reactor. The U_{rms} level in Figure 4.7 (c) is at its minimum at the catalytic wall and increases as the distance from the wall increases. Notably, the U_{rms} level is low at the reactor core and in a distinct region between the reactor core and the wall. However, to better understand the impact of reacting flow on the flow field, a non-reacting case under isothermal conditions, with the same inflow Reynolds number, is also simulated. Figure 4.7 (d)-(i) and Figure 4.8 (a)-(f) presents the transverse profiles of the mean velocity and its fluctuations at five different axial locations for reacting and non-reacting case, respectively. As shown in Figure 4.7 (d) for U_m in reacting flow, there is a characteristic overshooting of fast fluids near the wall. Consequently, U_m is at a high level near the wall and decreases to a low level at the reactor centre. As the downstream distance (x) increases, the U_m rises significantly compared



Figure 4.8 Case 3 (non-reacting): Transverse profiles of time-averaged (a) U_m (b) k (c) U_{rms} (d) V_{rms} (e) W_{rms} and (f) R_{UV} (or $U_{rms}V_{rms}$) at five streamwise locations. The number from 1 to 5 indicates axial locations shown in *Figure 4.7*(a).

to the non-reacting case, as shown in Figure 4.8 (a), due to the heat release from the catalytic wall. Again, the peak overshoot moves towards the reactor centre with increasing x_i indicating the development of a growing boundary layer. This type of velocity profile shows the effect of flow laminarisation reported in the literature [178]. To illustrate the laminarisation effect, the turbulent kinetic energy (k) for the reacting and non-reacting cases is compared at the same locations in Figure 4.7 (e) and Figure 4.8 (b), respectively. As shown in Figure 4.8 (b) for the non-reacting case, k exhibits peaks near the wall and maintains the similar levels across different axial locations, though k at the reactor core decreases as xincreases. In contrast, for the reacting flow in Figure 4.7 (e), k is low near the wall but high at the centre at x/H=3.0. Initially, k near the wall decreases and then increases with x, indicating an intensification of turbulence. However, the k values near the wall are significantly lower compared to those in the non-reacting case in Figure 4.8 (b), although the values at the reactor centre remain relatively consistent. This observation aligns with the previous findings in catalytically turbulent reacting flows in channels [114,116]. The characteristics of laminarising the flow and intensifying the turbulence near the wall are described as a two-stage process [113,179]. For instance, near the catalytic walls, heating induces local volumetric expansion, which enhances the ejection of low-speed hot fluid away from the walls. Initially, this effect is minor compared to the viscosity's impact on turbulence, resulting in laminarisation of the flow near the wall. However, as the distance x increases, this effect begins to counterbalance the viscosity's damping influence on turbulence, leading to an increase in k. Moving towards the reactor core, the ejection motion of hot fluid strengthens compared to the viscosity effect, resulting in a balance that slightly increases streamwise turbulence. Again, k exhibits two peaks near the reactor core in locations far downstream. This occurs due to the mixing of hot and cold fluids within the developing boundary layer. As described in [179], this mixing is driven by the ejection motion of hot fluids toward the reactor core and the sweep motion of cold fluids toward the wall, with the former dominating, leading to the observed downstream peaks. The transverse profiles of fluctuating components at these locations are presented in Figure 4.7 (f)-(i). As previously discussed, due to the same reasons, the U_{rms} shows a similar trend to k in Figure 4.7 (f), with near-wall values being lower compared to those in the non-reacting case shown in Figure 4.8 (c). Both V_{rms} and W_{rms} in Figure 4.7 (g)-(h) have small values near the wall and large values at the reactor core, and they decrease with x [29,30]. In comparison, the non-reacting case in Figure 4.8 (d)-(e) shows no significant variation in these quantities. However, the overall impact of velocity fluctuations throughout the reactor can be observed in the Reynolds stress (R_{UV}) profiles shown in Figure 4.7 (i). The R_{UV} exhibits peaks with both positive and negative values that correlate with the sign and magnitude of the velocity gradient [178]. The R_{UV} has the opposite sign to the velocity gradient and vanishes where the velocity gradient is zero. As reported in [178,180,181], the R_{UV} peak occurs at locations of peak U_{rms} and the R_{UV} value is zero at locations where U_{rms} is at a trough. The R_{UV} values near the wall in the reacting case are similar to those in the non-reacting case, as shown in Figure 4.8 (f). However, the R_{UV} peaks in reacting flow are higher near the reactor core, confirming the process of turbulent intensification caused by the ejection of hot fluids toward the reactor core.

Figure 4.9 illustrates the temperature and species distributions for the planar reactor. As shown in Figure 4.9 (a), the hot catalytic surfaces transfer heat to both the fluid and the solid. The conducted heat in the solid is directed towards the inert region near the inlet and outlet, preheating the incoming fresh mixture and the exiting combustion product, respectively. The fluid temperatures are high near the walls and low in the reactor core. The transverse profiles of the mean temperature (T_m) and its fluctuations (T_{rms}) are shown in Figure 4.9 (e)-(f) at different axial locations and, are symmetric about the reactor midplane on the y-axis. The results indicate that the cylinder core temperature remains below 400 K. However, unlike velocity fluctuations, T_{rms} in Figure 4.9 (f) increases with two peaks shifting towards the reactor core as x increases. This noteworthy aspect is explored based on the DNS predictions of Bae et al. [181], which consider classical heat transfer in a channel flow case. Their findings indicate that the wall heating reduces velocity fluctuations but has no significant effect on the T_{rms} predictions. As mentioned in their report, the underlying reason is that the enthalpy fluctuation remains at a considerable level downstream with the flow (for further details see [181]). Therefore, there seems to be no evidence supporting the notion that the effects of heating on the velocity and temperature fluctuations in turbulent flows are similar.

The mass fraction distribution of H_2 for the catalytic planar reactor is presented in Figure 4.9 (b). The H_2 concentration is high near the inert walls before entering the catalytic section. This is because of the high-affinity H_2 towards the hot walls reported by Mondal *et al.* [150]. As expected, due to the catalytic process, H_2 is consumed on the catalytic surfaces, leaving the reactor core unburned. Figure 4.9 (c) shows the distribution of instantaneous H_2O production in the catalytic planar reactor. The concentration of H_2O is high in the area where catalytic combustion occurs. Under the fuel-lean conditions, as discussed in a previous article [150], the mode of combustion here is mainly catalytic. However, the instantaneous contour of OH is presented in Figure 4.9 (d) to further investigate the role of homogeneous

(gas phase) combustion. The Y_{OH} scale is kept low, up to 2×10^{-5} , to capture the onset of homogeneous combustion. Since the catalyst produces less OH, the relatively low Y_{OH} levels on the catalytic surfaces is an indication of weak combustion of the gas mixtures.



Figure 4.9 Case 3 (reacting): Instantaneous contour of (a) T (b) Y_{H2} (c) Y_{H2O} (d) Y_{OH} onto the *xy*-plane at z/H = 0.75; Transverse profile of (e) T_m (f) T_{rms} (g) $Y_{H2,m}$ (h) $Y_{H2,rms}$, the number from 1 to 5 indicates axial locations shown in Figure 4.7(a).

Additionally, this low OH levels may result from the catalyst consuming OH produced by homogeneous combustion, thus inhibiting the combustion process. [150]. To provide further clarity, the streamwise profiles of the locally averaged (upper and lower surfaces) catalytic (C) and the integrated gas-phase (G) H₂ conversion rates are illustrated in Figure 4.10 (a). The results demonstrate that gas-phase conversion rates are significantly lower than catalytic rates, and both catalytic and homogeneous combustion can occur simultaneously at the same

location. Following the catalytic sections, the Y_{OH} levels result from the growth of the OH boundary layer near the hot inert wall with the flow.

Figure 4.9 (g)-(h) presents the H₂ transverse profiles of time-averaged mean and fluctuations at different axial locations. Figure 4.9 The $Y_{H2,m}$ values decrease as x increases. The maximum $Y_{H2,m}$ value near the end of the catalytic section is above 0.0038. Beyond that point, the $Y_{H2,m}$ values decrease in the reactor core and increase near the inert wall because of the characteristics of H₂ towards the hot walls. Like the temperature fluctuations (T_{rms}), the $Y_{H2,rms}$ in Figure 4.9 (h) has two peaks which increase with x and shift towards the reactor core. Similar observations of $Y_{H2,rms}$ were reported in an experimental work on catalytic combustion of hydrogen in a channel [84]. This indicates that the wall heating does not suppress the H₂ fluctuations. After the catalytic section at x/H=9, the $Y_{H2,rms}$ values increase significantly near the inert walls due to H₂ distribution while the $Y_{H2,rms}$ peak values remain similar in the reactor core.

To assess the heat and H_2 mass exchange between the flow and catalytic surfaces, the Nusselt number (*Nu*) and Sherwood number (*Sh*) of H_2 for the planar reactor are calculated on the reactor inner surface and shown in Figure 4.10 (b). The presented *Nu* and *Sh* values are the averages of those from the upper and lower surfaces of the planar reactor. Their definitions are given as follows:

Nusselt Number:
$$Nu = \frac{h_c(2H)}{\lambda_g}$$
 where $h_c = \frac{\lambda_g \frac{\partial T}{\partial y}|_{wall}}{T_w - T_{ref}}$, (4.45)

where h_c is the heat transfer coefficient, λ_g is the fluid conductivity and $\partial T/\partial y$ is the gradient of temperature at the solid surface. The T_w and T_{ref} are the wall surface and the reference temperatures, respectively. For convenience, T_{ref} is the average of T_{in} and T_{adb} where T_{adb} is the adiabatic temperature of H₂/air mixture. In Figure 4.10 (b), Nu is high at the entrance of the reactor and gradually decreases along the length of the reactor. This is because the thermal boundary layer is thin at the entrance, causing a high temperature gradient and a high value of Nu. As the fluid flows along the reactor length, the thermal boundary layer thickens and the temperature gradient decreases, resulting in a decrease in Nu.

Sherwood Number:
$$Sh = \frac{k_c(2H)}{D_m}$$
 where $k_c = \frac{D_m \frac{\partial C_{H2}}{\partial y}}{C_{H2,w} - C_{H2,ref}}$ (4.46)

where k_c and D_m are the H₂ mass transfer coefficient and mass diffusivity, respectively. $\partial C_{H2}/\partial y$ is the gradient of mean H₂ mass concentration at surface. $C_{H2,w}$ and $C_{H2,ref}$ are the surface and the reference concentration of H₂, respectively. $C_{H2,ref}$ is considered as an average of concentrations at T_{in} , and at T_{adb} where H₂ is considered to be completely consumed.



Figure 4.10 Case 3 (reacting): (a) The predicted locally averaged (upper and lower surfaces) catalytic conversions (*C*) and the integrated (over the reactor flow height) gaseous conversions (*G*) along the streamwise direction. (b) Local distribution of Nu, Sh, τ_w and T_w on reactor inner surface.

As shown in Figure 4.10 (b), the *Sh* is high at the beginning of the channel due to a high H₂ concentration gradient caused by thermal diffusion. This value gradually decreases as the concentration boundary layer develops in the inert region. The boundary layer disruption, caused by catalytic combustion, results in a high *Sh* at the beginning of the catalytic section, which then significantly drops up to x = 0.01. Beyond this point, the variation in *Sh* remains insignificant with increasing x up to the end of the catalytic section. Initially, the H₂ concentration rate at the catalytic surface is high, resulting in a steep H₂ concentration gradient and a high *Sh* value. As x increases, the concentration level becomes insufficient for effective catalytic combustion, leading to a decreased concentration near the wall increases due to the standard Fickian diffusion, causing a further decrease in the H₂

concentration gradient, and thus, resulting in an insignificant effect of *Sh* in the inert region. However, the *Nu* and *Sh* in Figure 4.10 (b), exhibit different behaviours, indicating that the typical mass and heat transfer analogy does not apply in this case due to catalytic combustion affecting the wall temperatures and fluid properties. Additionally, the wall shear stress (τ_w) along the reactor is shown in Figure 4.10 (b). τ_w is initially high at the entrance due to the developing boundary layer, and then it gradually decreases. This decrease is attributed to increased viscosity caused by heat transfer from the wall. Overall, the behaviours of Sherwood and Nusselt number and the shear stress appear to be quite different. Hence, considering the catalytic process, the general analogy among momentum, heat, and mass transfer does not seem to be totally applicable.

Figure 4.11 (a)-(c) illustrates the flow field of the non-planar catalytic reactor. The contours of the instantaneous x-velocity (U) and mean x-velocity (U_m) in Figure 4.11 (a)-(b) reveal the presence of a recirculation zone across the domain, attributed to the flow restriction caused by the cylindrical rods. This recirculation occurs predominantly at the back of the cylinders. The flow velocity rises above and below the cylinders, resulting in increased streamwise velocity fluctuations (U_{rms}) shown in Figure 4.11 (c). Both the U_m and U_{rms} velocity fields are relatively symmetric about the reactor midplane on the y-axis. The strongest magnitudes of U_{rms} are located above and below full cylinder 3 and its neighbouring half-cylinders. This is due to the adjacent heated walls resulting from the catalytic combustion causing fluid expansion. To better understand this effect, the transverse distribution of mean axial velocity and turbulent fluctuations at different axial locations are shown in Figure 4.11 (d)-(h). The locations are considered downstream of full cylinders, indicated by the vertical lines and numbered from 1 to 6 in Figure 4.11 (a). As shown in Figure 4.11 (d), the peak of U_m occurs between the half and full cylinders, while the troughs are located at the back of the full cylinders, which are wake zones due to flow separation. The U_m peak values at location 2 increase significantly and become, nearly 1.5 times compared to location 1. At locations 2 and 3, the peak values are similar but increase again at locations 4 and 5. Subsequently, the peak values at locations 5 and 6 remain similar and become, nearly 1.85 times compared to location 1. As mentioned earlier, the heat transfer form hot catalytic surfaces to fluid here is strong causing fluid expansion and high streamwise velocity. This also significantly affects the velocity fluctuations (U_{rms} , V_{rms} and W_{rms}) shown in Figure 4.11 (f)-(i). The fluctuations successively increase at locations 2 and 3 compared to 1. After that, the variations at locations 4, 5, and 6 become smaller. However,

99

to analyse the laminarisation effect, the turbulent kinetic energy (k) is presented in Figure 4.11 (e).



Figure 4.11 Case 4 (reacting): Contours of (a) instantaneous x-velocity, U (b) timeaveraged mean x-velocity, U_m and (c) its fluctuations, U_{rms} onto the xy-plane at z/H = 0.75 Transverse profiles of time-averaged (d) U_m (e) k (f) U_{rms} (g) V_{rms} (h) W_{rms} and (i) R_{UV} (or $U_{rms}V_{rms}$) at six streamwise locations.

Unlike that in the planar reactor, the value of k increases with streamwise locations, indicating a strengthening of turbulent flow. Furthermore, the resolved Reynolds stress (R_{UV}) variations at the same locations are shown in Figure 4.11 (i). Similar to the velocity fluctuations and k, the R_{UV} peaks also increase with streamwise locations, confirming the intensified turbulent effects in the nonplanar reactor.

Figure 4.12 (a)-(d) illustrates the contour of instantaneous temperature and species mass fraction for the non-planar reactor. As shown in Figure 4.12 (a), the fluid temperatures are high near the walls and cold far from the walls up to the 4th full cylinder. As effective heat transfer occurs from the wall to the fluid, the temperature variations diminish significantly, leading to nearly uniform temperatures far downstream. The transverse profile of the mean temperature (T_m) and the temperature fluctuations (T_{rms}) for the non-planar reactor are presented in Figure 2.11(e) and Figure 4.12 (f), respectively. As discussed earlier, T_m values are high near the walls due to heating provided by the catalytic walls and minimum far from walls. The T_m increases significantly up to location 5 and beyond that, the variation between locations 5 and 6 becomes smaller, indicating a weakening of the catalytic combustion process though temepature by itself is insufficient for judging the strength of catalytic reactions. In Figure 4.12 (f), the T_{rms} has peaks in the regions between half and full cylinders. The T_{rms} peak values exceed 80 K and are highest at locations 2 and 3. An interesting observation is that T_{rms} , compared to the planar reactor, exhibit low values at far downstream locations, even though the velocity fluctuations are high. This observation reaffirms the minimal impact of wall heating on temperature fluctuations, as discussed earlier in Figure 4.9 (f) for planar reactor. It also emphasises that the enthalpy fluctuation as reported in article Bae *et al.* [181] plays a significant role in temperature fluctuations.

The mass fraction of H₂ shown in Figure 4.12 (b) decreases along the streamwise direction in a non-planar reactor, due to catalytic combustion on the Pt-coated cylindrical surfaces. Compared to planar reactor, H₂ consumption is more effective, owing to the increased mass transfer of H₂ with the flow towards the catalytic surfaces. To gain a better understanding, the mean and fluctuation of the mass fraction of H₂ transverse distribution at six axial locations are presented in Figure 4.12 (g) and Figure 4.12 (h), respectively. These axial locations are shown in Figure 4.11. The $Y_{H2,m}$ values for non-planar reactor, shown in Figure 4.12 (g), significantly decrease up to location 5 and beyond that, the variations become smaller. Similar behaviour is also observed in the temperature profile discussed in Figure 4.12 (e). This is due to insufficient H₂ concentration available for the catalytic process. The maximum $Y_{H2,m}$ value at the location 6 drops below 0.0012. The $Y_{H2,rms}$ in Figure 4.12 (h)



Figure 4.12 Case 4 (reacting): Instantaneous contour of (a) T (b) Y_{H2} (c) Y_{H2O} (d) Y_{OH} onto the *xy*-plane at z/H = 0.75; Transverse profile of (e) T_m (f) T_{rms} (g) $Y_{H2,m}$ (h) $Y_{H2,rms}$, the number from 1 to 6 indicates axial locations shown in Figure 4.11(a) (i) The predicted locally averaged (upper and lower cylindrical surfaces) catalytic conversions (*C*) and the integrated (over the reactor flow height) gaseous conversions (*G*) along the streamwise direction.

has a nearly identical profile to T_{rms} with peaks in the regions between half and full cylinders. The $Y_{H2,rms}$ peak values are highest at locations 2 and 3 shown in Figure 4.12 (h). After these locations, the $Y_{H2,rms}$ peak values gradually decrease with increasing x. These observations of the non-planar reactor, along with the results for the planar reactor in Figure 4.9 (f) and (h), indicate that H₂ mass fraction fluctuations are strongly linked to temperature fluctuations. In contrast to the planar reactor, the combustion process in the non-planar reactor is entirely catalytic, as indicated by the absence of significant OH throughout the domain, as seen in Figure 4.12 (c). For further confirmation, the locally averaged catalytic (*C*) and the integrated gas-phase (*G*) H₂ conversion rates are presented in Figure 4.12 (i). The flat profile with zero conversion rates indicates the absence of homogeneous combustion under the current operating conditions. Figure 4.12 (d) shows the instantaneously produced H₂O distribution resulting from catalytic reactions within the reactor. The H₂O concentration increases along the length of the reactor, with the variation becoming smaller after $x\approx 0.018$. This is because the low concentration of H₂ limits further catalytic combustion.

However, the distribution of Nu and Sh of H₂ species over the catalytic surfaces along the length of the non-planar reactor is shown in Figure 4.13 (a) and Figure 4.13 (b), respectively. Due to symmetry, both Nu and Sh are averaged from the values of the upper and lower cylindrical surfaces. As expected, close to the stagnation points of the cylinders where the boundary layers are thin, Nu and Sh values are high. The opposite applies to the wake region at the back of cylinders. This is why, the quarter portion at the back of the cylinders is uncoated. However, both the Nu and Sh peaks occur in most of the cylinders are high compared to downstream cylinders, indicating more effective catalytic processes in these regions. As a result, significant variations in turbulent quantities discussed earlier are observed in those regions. However, the wall shear stress distribution (τ_w) over the cylindrical surfaces is shown in Figure 4.13 (c) to relate the overall impact of flow on heat and mass transfer. As seen in the plot, the τ_w significantly increases from the stagnation point because of the high velocity gradient and becomes a peak at a similar location of Nu peak. After that, the τ_w decreases as velocity gradient decreases.



Figure 4.13 Case 4 (reacting): local distribution of (a) Nu, (b) Sh and (c) τ_w on catalytic cylindrical surfaces.

Figure 4.14 shows the hydrogen conversion along the reactor length. Conversion values are obtained using the following formula:

$$H_{2} \operatorname{conversion}(\%) = \frac{Y_{H2,avg,in} - Y_{H2,avg,x}}{Y_{H2,avg,in}} \times 100$$
(4.47)

where $Y_{H2,avg,in}$ and $Y_{H2,avg,x}$ are the average H₂ mass fraction at the inlet and axial positions, respectively. The $Y_{H2,avg}$ values are estimated at streamwise locations taking into account the spatially averaged transverse and spanwise directions. With the same amount of catalytic surface area, the non-planar reactor achieves a significantly higher H₂ conversion rate. Although the residence time may slightly differ between the two reactors, this conversion rate remains a useful metric for comparing their effectiveness. As previously discussed, the cylindrical rods placed across the flow in the non-planar reactor significantly enhance the mass transfer of H₂ towards the catalytic surface, thereby improving the catalytic combustion process. Notably, the first ~60% H₂ conversion is achieved using only 50% of the catalyst, whereas the remaining catalyst contributes to an additional ~20% H₂ conversion. In contrast, the planar reactor achieves ~37% and ~11% H₂ conversion for the first and second 50% of the catalyst, respectively. Interestingly, the initial 37.5% of the catalyst in the non-planar reactor.

The non-planar reactor, while advantageous, experiences a significant pressure drop of 97.2%, compared to 48.7% in the planar reactor. This drop, defined as the percentage decrease from the inlet pressure, results from the flow separation and constriction caused by the cylinders placed inside the reactor. In the catalytic reactor design, balancing the H₂



Figure 4.14 Hydrogen conversion (%) along the length of the reactor.

conversion efficiency with the pressure drop is crucial, impacting throughput, energy efficiency, and costs. Future work should focus on the further optimisation of reactor geometry to improve flow distribution as well as the refining catalyst coating strategies to achieve this balance.

4.5 Discussion

Catalytic hydrogen combustion in a planar type reactor has been studied for many years. However, the use of staggered cylinders with catalyst coatings in a non-planar reactor to enhance the catalytic performance has not been thoroughly explored prior to this study. The cylinders intensify turbulence, which further improves the mass and heat transfer at the surfaces, leading to an increased fuel conversion. To model turbulence, LES is an attractive option; however, due to its high computational demands, 3D modelling with LES in such reactors has been scarcely explored in the literature. Hence, this paper presents LES modelling in a non-planar reactor to investigate the intensification of the catalytic process. The results demonstrate that the LES model effectively predicts the catalytic reacting flow. A non-planar reactor with cylindrical surfaces coated with catalysts is studied and compared with a planar reactor having the same inner surface coating. The turbulence in the non-planar reactor significantly enhances mass and heat transfer at the catalytic surfaces, thereby intensifying the catalytic process. Under the considered operating conditions in the nonplanar reactor, gas-phase combustion is completely inhibited, and the catalytic surface temperatures are comparatively low. Furthermore, H₂ conversion in the non-planar reactor is significantly higher than in the planar reactor. The increased catalytic activity combined with lower catalytic surface temperatures provides a basis for optimising catalytic reactor design for various applications.

4.6 Conclusions

The turbulent catalytic combustion of a highly lean, premixed fuel-lean hydrogen/air mixture with an equivalence ratio of 0.15 over a platinum catalyst, at an inflow temperature of 300 K and a Reynolds number of 3500, was investigated. This study examined both the planar and non-planar reactors using large eddy simulation. The numerical method relied on the structured finite volume discretisation, incorporating the turbulent kinetic energy sub-grid model for LES and the eddy dissipation concept for computing species reaction rates. Before analysis, the numerical method underwent validation for both configurations, with experimental results utilised for comparison. The key findings are summarised as follows.

- In the planar catalytic reactor, the flow turbulence remains nearly consistent along the reactor length.
- The non-planar reactor with staggered cylindrical rods significantly increases turbulence, enhancing heat and mass exchange with the catalytic surface and thereby intensifying catalytic combustion.
- In the planar reactor, both *Nu* and *Sh* decrease along the reactor length, while, in nonplanar reactor, *Nu* and *Sh* are most effective on both the full and half cylindrical coated surfaces.
- In contrast to the planar reactor, hydrogen conversion in the non-planar reactor is purely catalytic and occurs at lower surface temperatures.
- The non-planar reactor improves H₂ conversion to 80.04% at the reactor exit, compared to 48.1% in the planar reactor.
- The non-planar reactor achieves the same output as the planar reactor while using 62.5% less catalyst.

However, the catalyst quality and loading might have significance influence on catalytic performance and has not investigated in this chapter due to challenges for numerical modelling of catalyst content and loading. Therefore, an experimental study in next chapter is carried out to analyse the effect of catalyst quantity on the catalytic process.

Chapter 5: Effect of catalyst quantity on catalytic combustion

Abstract

Catalytic combustion of ultra-lean hydrogen/air mixture is a promising technique for generating power with zero emissions, especially in low-temperature heating applications. A major challenge of this technique is the reliance on noble catalysts, which are both expensive and rare in nature. Therefore, minimising the use of catalysts is essential for costeffective catalytic combustion system designs. This work experimentally investigates the effects of varying catalyst contents and loadings in a packed bed tubular catalytic reactor. Active catalyst sites are applied to the surfaces of Al₂O₃ pellets that make up the packed bed. An ultra-lean premixed mixture of 2% H₂ in the air is used for the catalytic combustion. The catalyst contents tested include 0.3%, 0.5%, and 1.0% Pt in Pt/Al₂O₃ pellets, and 0.5% and 5.0% Pd in Pd/Al₂O₃ pellets. Catalyst loadings for both catalyst pellets were set at 1.0 g, 2.5 g, and 5.0 g. Measurements were taken in the packed bed reactor across the flow rates ranging from 1 LPM to 5 LPM. The results show that the packed bed with higher Pt or Pd content generates elevated combustion temperatures and demonstrates an effective catalytic performance. Additionally, the occurrence of super-adiabatic conditions was observed, and hydrogen conversion rates were significantly influenced by the catalyst contents. Notably, the pellets with high Pt or Pd content, exhibited catalytic performance comparable to higher catalyst loadings at different flow conditions, even with a loading of 1 g at low flow rates.

5.1 Introduction

In the previous studies (Chapters 2-4), we explored a catalytic monolithic reactor operating near the lower flammability limit of hydrogen/air mixture, aiming to intensify the catalytic process by incorporating an array of staggered catalyst-coated cylinders. This design significantly improved the catalytic performance compared to a traditional monolithic reactor without cylinders, primarily due to an improved mass and heat convection toward the catalyst surface of the burner. Although these studies demonstrated an improved catalytic performance, further investigation is essential regarding the minimum catalyst usage required for cost-effective catalytic combustion system designs.

However, numerical investigations in earlier chapters were favoured over experimental methods due to the challenges of obtaining accurate measurements near catalytic surfaces. Moreover, numerical modelling offered a convenient approach to exploring the details of catalytic reacting flows within monolith reactors and the intensification of the CHC process over catalytic surfaces. However, addressing the effect of catalyst loading to minimise the use of expensive catalysts remains a significant challenge in numerical modelling. This difficulty arises because the chemical kinetics involved in predicting the CHC process are highly sensitive to the catalyst type and vary with changes in catalyst loading. Therefore, this study focuses on the experimental investigation of a catalytic reactor to analyse the effect of catalyst quantity on the CHC process.

While minimising catalyst usage is vital for expanding the application of catalytic hydrogen combustion (CHC) process, the design constraints of constructing a monolithic reactor with staggered cylinders led us to use a packed bed reactor [59,182,183] instead. Both the reactor types exhibit similar catalytic behaviour along the reactor length, making the packed bed reactor a simpler and more practical option for experimental investigation. A typical example of monolithic catalytic reactor and catalyst-packed reactor application is shown in Figure 5.1 (a) and (b), respectively. Nonetheless, the results from the packed bed reactor remain useful for predicting the catalytic performance of a monolithic reactor with catalyst-coated cylinders. Because both reactors operate on a premixed principle. Moreover, the catalytic conversion of reactants occurs in both reactors while flowing over the catalyst-coated surfaces.



109



staggered catalyst coated cylinders inside monolith

(b) Catalytic packed bed reactor [185]



Figure 5.1 (a) monolithic catalytic reactor (b) catalyst packed bed reactor. Figures from [184,185] are reused with permission.

(a) Catalytic monolith reactor [184]

5.2 Experiments

5.2.1 Experimental setup and equipment

The experiment investigates catalytic hydrogen combustion in a packed bed reactor using various catalyst loadings. The experimental setup, illustrated in Figure 5.2, consists of a regulated inflow system with a flame arrestor, a U-tube quartz reactor containing the packed bed of catalyst-coated alumina pellets, a diffuser glass tube attached to the reactor's exit to reduce flow velocity and aid in sample acquisition for the mass spectrometer, and a suite of detection instruments for measurement. The tube quartz reactor is 13 mm in diameter and 1 mm in thickness and is connected to inlet steel tubing. A hydrogen/air mixture (2% H₂) is supplied from a premixed cylinder and a mass flow controller is used to regulate flow at 1 atm into the quartz reactor. Catalyst pellets are loaded inside the quartz reactor and placed at a particular position supported by a steel wire mesh. Another steel wire mesh is placed at the reactor exit for uniform flow of the combustion product. A gas sample probe is positioned at the centre of the diffuser glass, from which the gas sample is drawn into a mass spectrometer to measure the concentration of H₂. The temperature of the catalyst is measured using a N-type thermocouple. The thermocouple probe tip is placed at the centre location of the



Figure 5.2 Schematic of experimental setup

tube reactor and positioned while measuring the temperature at the bottom, middle and top of the catalyst-packed bed. Moreover, an IR camera is placed at a distance of 25 cm from the reactor to capture the temperature distribution on the quartz tube wall.

5.2.2 Instrumentation

5.2.2.1 Temperature measurement

An N-type thermocouple is used to measure the temperature of the catalytic bed, positioned at the desired location. The thermocouple's output is recorded using a Picolog-TC08 temperature data logger and stored on a computer hard drive.

Since the hydrogen combustion process is colourless, an infrared thermal camera is used to monitor and visualise the temperature distribution on the quartz wall of the reactor. The IR camera lens is positioned at a distance of 25 cm from the reactor.

5.2.2.2 Mass spectrometry (MS)

Hydrogen gas detection is carried out using a quadrupole mass spectrometer (QMS) (European Spectrometry Systems Ltd. GeneSys Evolution QMS400). The QMS consists of four key components: an ioniser, where electrons from a heated filament bombard the gas sample to ionise the molecules; an ion accelerator, which propels the ions; a mass filter, comprised of four parallel metal rods, that selectively filters ions based on their mass-to-charge (m/z) ratio; and a detector that collects the filtered ions. Only ions with a specific m/z ratio, corresponding to a given voltage ratio, reach the detector, while ions with unstable trajectories are deflected and expelled after colliding with the rods. When an ion hits the detector, it generates an electrical current proportional to its abundance. This data is then processed by a computer to analyse the electrical current corresponding to the m/z ratios of the gas sample species.

5.2.2.3 Data calculation

The electrical current for gas species in the QMS is obtained using the Multiple Ion Detection (MID) mode. In MID mode, gas species such as N_2 , O_2 , CO, Ar, and H_2 are detected by measuring the electrical current corresponding to their respective m/z ratios. Since the fuel mixture in the current analysis consists of 2% H_2 in air, and there is no significant change in the air composition after the catalytic process, only the H_2 concentration is calculated to assess performance.

To ensure accuracy, a background subtraction is performed in MID mode. For this, the inlet of the QMS is closed to create a zero-gas condition, which allows for the subtraction of background signals. This step is essential for detecting low concentrations of species, particularly H₂ in this case. Following background subtraction, the MID mode is calibrated using the 2% H₂ in air mixture at 1 bar. The sensitivity of the QMS to H₂ (K_{H_2}) for the m/z = 2.0 signal is then calculated using the following equation.

$$K_{H2} = I_{H2,ref} / C_{H2,ref}$$
 (5.1)

where,
$$I_{H2,ref} = I_{H2,total} - I_{H2,res}$$
 (5.2)

Where $I_{H2,ref}$ is the reference current generated at the reference hydrogen concentration $C_{H2,ref}$, calculated by subtracting the residual current under zero-gas conditions from the total current. The hydrogen concentration under a particular operating condition (*x*) during the catalytic combustion process can be determined using the following equation.

$$C_{H2}(x) = I_{H2}(x) / K_{H2}$$
 (5.3)

where,
$$I_{H2}(x) = I_{H2,total}(x) - I_{H2,res}(x)$$
 (5.4)

5.3 Results and discussion

To assess the catalytic performance, the flow rate of the premixed hydrogen/air (2% H₂) mixture through the catalytic packed bed reactor is varied between 1 and 5 LPM at atmospheric pressure. The catalyst loadings in the catalyst bed are set at 1.0 g, 2.5 g, and 5.0 g. Two types of catalyst pellets (Pt/Al₂O₃ and Pd/Al₂O₃) are tested. Characteristics of pellets at various catalyst contents are detailed in the Table 5.1. The pellet sizes are $3mm \pm 0.2mm$. The characteristics of pellets at various catalyst contents are detailed in the Table 5.1. The pellet sizes are $3mm \pm 0.2mm$. The characteristics of pellets at various catalyst contents are detailed in the **Error! R eference source not found.**. The gas hourly space velocity (GHSV) for a 5.0 g catalyst is estimated for flow rates of 1, 2, 3, 4, and 5 LPM as follows: 13.2, 26.4, 39.6, 52.8, and 66 hr⁻¹, respectively. For catalyst bed weights of 1.0 g and 2.5 g, the GHSV values are 5 times and 2 times that of the 5.0 g bed, respectively.

Catalyst type	Supplier	Average pore radius (Å)	BET specific surface area (m²/g)	Total pore volume (cc/g)
0.3% Pt/Al ₂ O ₃	Thermo	47.3	118.22	0.2.79
	Scientific			
0.5% Pt/Al ₂ O ₃	Jhonson	53.3	92.716	0.247
	Matthey			
1.0% Pt/Al ₂ O ₃	Thermo	52.2	96.742	0.252
	Scientific			
0.5% Pd/Al ₂ O ₃	Alfa Easer	46.7	112.261	0.262
5.0% Pd/Al ₂ O ₃	Thermo	50.7	224.255	0.568
	Scientific			

Table 5.1 List of catalyst samples

5.3.1 Reactor temperatures

Figure 5.3-Figure 5.6 present reactor wall temperature maps of listed sample in Table 5.1 for various flow rates and catalyst loadings. Although the temperatures shown do not represent the exact inner wall temperatures due to emissivity losses in the quartz wall, the thermal images captured by the IR camera still provide valuable insights into the internal catalytic processes and heat transfer dynamics of the reacting flow.

In Figure 3(a), for the 0.5% Pt/Al₂O₃ catalyst with a 1.0 g loading, the wall temperatures at the catalyst location range from 55°C to 60°C and remain relatively constant across all flow rates. However, as the flow rate increases, the heated region of the wall expands, indicating enhanced convective heat transfer. When the catalyst loading increases to 2.5 g, as shown in Figure 3(b), the wall temperatures rise significantly, with peak values ranging between 80°C and 100°C. As expected, both the extent of the heated region and the wall temperatures increase with higher flow rates. Further increasing the catalyst loading to 5.0 g does not result in higher maximum wall temperatures compared to the 2.5 g case, but the heated region continues to expand with increasing flow rate. The larger heated area at higher catalyst loadings is attributed to the increased thermal mass of the additional catalyst.

For the 1.0% Pt/Al₂O₃ catalyst, as depicted in Figure 4, the wall temperatures are captured under the same inflow conditions and catalyst loadings like the 0.5% Pt/Al₂O₃ catalyst. The heated region at the catalyst location, along with the pattern of expanding heated areas with higher flow rates, is very similar to the 0.5% Pt/Al₂O₃ case. However, the wall temperatures for the 1.0% Pt/Al₂O₃ catalyst are consistently higher across all studied conditions, indicating

greater heat generation from the catalytic process. The increased catalytic activity of the 1.0% Pt/Al₂O₃ catalyst can be attributed to its higher platinum content. To further investigate the effect of catalyst content, 0.3% Pt/Al₂O₃ catalyst of similar characteristics as mentioned in Table 5.1 is tested under the same operating conditions as 0.5% Pt/Al₂O₃ and 1.0% Pt/Al₂O₃. Interestingly, no catalytic activity is observed with the 0.3% Pt/Al₂O₃ catalyst, likely due to its low platinum content.

Similarly, the wall temperature maps for 0.5%Pd/Al₂O₃ and 5%Pd/Al₂O₃ catalysts are presented at different catalyst loading in Figure 5.5-Figure 5.6. The results show that these catalysts are also effective for hydrogen oxidation and producing heat. However, the relative effect of catalysts on the catalytic combustion process and catalyst temperatures are discussed in the following section.

(a) Catalyst loading = 1.0 g



1 LPM 2 LPM 3 LPM 4.0 LPM 5.0 LPM

Figure 5.3 Reactor wall temperatures (°C) obtained from IR thermal camera, 0.5% Pt/Al_2O_3

(a) Catalyst loading = 1.0 g



Figure 5.4 Reactor wall temperatures (°C) obtained from IR thermal camera, 1.0% Pt/Al_2O_3

(a) Catalyst loading = 1.0 g



Figure 5.5 Reactor wall temperatures (°C) obtained from IR thermal camera, 0.5% Pd/Al_2O_3 $\,$



Figure 5.6 Reactor wall temperatures (°C) obtained from IR thermal camera, 5.0% Pd/Al₂O₃

To gain better understating of the catalyst temperatures during catalytic combustion process, a thermocouple is used and positioned for measuring temperatures at the bottom, middle, and top locations of the catalytic packed bed. As expected, measured catalyst temperatures

are significantly higher than the wall temperatures. Figure 5.7 and Figure 5.8 present the temperature profiles as a function of flow rates for the 0.5% Pt/Al₂O₃ and 1.0% Pt/Al₂O₃ catalysts, respectively. The vertical bar in both plots presents the measurement uncertainty. For the 0.5% Pt/Al₂O₃ catalyst with a 1 g loading, the temperature at the bottom of the bed decreases with increasing flow rates, while the temperature at the middle remains consistently high, around 140°C, across all flow rates. At the top of the bed, temperatures are about 15°C lower than at the middle but increase slightly with flow rate due to enhanced convective heat transfer. When the catalyst loading is increased to 2.5 g, the temperature at the bottom reaches a maximum at 2 LPM before decreasing with higher flow rates. The temperatures at the middle and top of the bed increase with flow rate, with a temperature difference of approximately 20°C between the two locations. Overall, the temperature values for the 2.5 g catalyst loading are higher than those for the 1 g loading due to the increased catalyst mass involved in the catalytic process. For the 5.0 g catalyst loading, the bottom location temperatures are higher than for the 2.5 g case but follow a similar trend with flow rates. The temperatures at the middle remain in a similar range as for the 2.5 g loading, while the top temperatures are comparatively lower, likely due to the thicker catalytic bed, which hinders heat dissipation with increased flow rates.

For the 1.0% Pt/Al₂O₃ catalyst, as shown in Figure 5.8, the temperature profiles under similar inflow and catalyst loading conditions are like the 0.5% Pt/Al₂O₃ catalyst. However, the overall temperatures are higher, indicating increased heat generation from the catalytic process due to the higher platinum content. Unlike the 0.5% Pt/Al₂O₃ catalyst, the maximum temperatures at the bottom of the bed for the 1.0% Pt/Al₂O₃ catalyst are observed for both the 2.5 g and 5.0 g loadings. Interestingly, in both cases, the temperatures are experienced above the adiabatic temperatures (~178°C). Interestingly, in both cases, the observed temperatures exceed the adiabatic limit (~178°C) at the front location of the catalyst bed, highlighting the super adiabatic nature of the catalytic process, which is attributed to the low Lewis number of the hydrogen/air mixture [53,66,129]. Additionally, the temperatures at the middle and top of the bed increase with flow rates, with temperature differences between the locations of approximately 20°C and 40°C for 2.5 g and 5.0 g loadings, respectively.





(a) 1g













Figure 5.7 Catalyst (0.5% Pt/Al₂O₃) bed temperatures at different locations for loading: (a) 1g (b) 2.5g (c) 5g

Figure 5.8 Catalyst (1% Pt/Al₂O₃) bed temperatures at different locations for loading: (a) 1g (b) 2.5g (c) 5g



Figure 5.9 Catalyst (0.5% Pd/Al₂O₃) bed temperatures at different locations for loading: (a) 1g (b) 2.5g (c) 5g



Figure 5.10 Catalyst (5% Pd/Al₂O₃) bed temperatures at different locations for loading: (a) 1g (b) 2.5g

Similarly, the profile of catalyst bed temperatures for 0.5% Pd/Al₂O₃ and 5.0% Pd/Al₂O₃ at different locations are illustrated in Figure 5.9 and **Error! Reference source not found.**, respectively. The trend of the profiles with flowrate is similar as experienced for 0.5% Pt/Al₂O₃ and 1.0% Pt/Al₂O₃ in Figure 5.7 and Figure 5.8. In addition, the super adiabatic temperatures are observed for 5.0% Pd/Al₂O₃ catalysts at the front location of the catalyst bed while operating at higher flow rates. This indicates that high catalyst content enhances the catalytic process and leads to super adiabatic temperature. However, to have a better understanding of the catalytic process, the effect of catalyst content and various catalyst loading on the catalytic conversion rates are discussed in the following section.

Details of the temperature measurements for 0.5% Pt/Al₂O₃,1.0% Pt/Al₂O₃, 0.5% Pd/Al₂O₃ and 5.0% Pd/Al₂O₃ at various catalyst loadings are presented in Table 5.3, Table 5.3, Table 5.4 and Table 5.5, respectively. The uncertainty calculated here is based on the population standard deviation.

Table 5.2 Thermocouple measurements (°C) for catalysts 0.5% Pt/Al ₂ O ₃							
Loading	Probe location	LPM	Test-1	Test-2	Test-3	Mean	Error
	front	1	122.01	118.05	125.02	121.69	1.656
		2	112.04	109.08	115	112.04	1.414
		3	104.84	101.02	108.40	104.75	1.655
		4	98	101.4	95.9	98.43	1.308
		5	92.17	95.09	88.28	91.84	1.656
		1	131	135	126.1	130.7	3.634
		2	142.38	145.59	138.47	142.14	2.867
1g	mid	3	143.12	145.24	140.03	142.79	2.058
		4	140.40	143.49	136.51	140.09	2.867
		5	137.04	142.05	135.01	138.33	2.867
			110.4	100 7	111 51	110 5	0.1.00
	back	1	113.4	109.5	114.61	112.5	2.160
		2	121.94	125.85	118.91	122.23	2.867
		3	123.49	124.70	126.58	124.92	1.247
		4	126.86	128.9	123.37	126.36	2.254
		5	128.25	131.75	133.5	131.16	2.054
		1	120.52	100 50	104 67	107.00	1 4 4 0
2.5g	front	1	128.63	130.56	124.67	127.93	1.440
		2	145.95	148.85	141.76	145.52	1.655
		3	143.32	140.51	146.41	143.42	1.414
		4	133.8	137.84	130.76	134.13	1.655
		5	120.47	125.27	117.36	121.03	1.905
	mid	1	110.78	114.85	108.71	111.44	2.494
		2	144.8	147.73	141.87	144.8	2.449
		3	157.20	153.34	159.27	156.60	2.494
		4	161.51	164.71	158.31	161.51	2.449
		5	162.35	165.65	160.95	162.98	2.054
		·	1				
		1	90.53	93.32	87.44	90.43	2.450
		2	115.77	112.63	117.7	115.3	2.054

	back	3	137.92	138.32	137.02	136.7	1.247	
		4	141.60	145.54	142.78	143.30	1.699	
		5	144.91	147.83	140.73	144.49	2.867	
	front	1	131.7	130.5	128.65	130.3	0.740	
		2	153.63	148.86	149.95	150.78	1.186	
		3	154.62	149.54	155.32	153.16	1.537	
		4	149.54	147.7	151.9	149.71	0.945	
		5	141.55	143.33	139.37	141.36	0.942	
5g		1	101.43	104.71	100.78	102.33	1.752	
	mid	2	140.9	143.6	141.8	142.16	1.211	
		3	152.7	153.29	152.25	152.74	0.409	
		4	156.84	154.63	158.41	156.62	1.640	
		5	157.23	155.65	160.69	157.84	2.086	
	back	1	50.41	53.47	57.53	53.76	2.867	
		2	74.97	72.65	77.72	75.12	2.044	
		3	101.52	104.02	100.22	101.92	1.657	
		4	117.39	115.73	112.54	115.22	1.961	
		5	129.45	127.73	131.84	129.73	1.642	

Table 5.3 Thermocouple measurements (°C) for catalysts 1% Pt/Al ₂ O ₃											
Loading	Probe location	LPM	Test-1	Test-2	Test-3	Mean	Error				
		1	141.18	144.29	140.10	141.85	0.9813				
	front	2	149.73	152.83	147.93	150.16	1.183				
		3	148.84	146.90	151.97	148.94	1.414				
		4	143.32	145.44	140.50	143.08	1.1863				
		5	138.53	140.62	136.74	138.63	0.942				
		1	138 771	140.51	1/1 661	140.31	1 1 8 5				
		2	130.//1	161.94	141.001	164.57	2.245				
1		2	164.54	101.84	167.34	164.57	2.245				
Ig	mid	3	171.58	174.48	169.28	171.78	2.127				
		4	175.68	172.88	176.26	174.94	1.475				
		5	175.29	176.19	174.4	175.29	0.730				
	back	1	115.82	113.94	118.73	116.16	1.970				
		2	142.75	141.84	144.96	143.18	1.310				
		3	150.7	153.64	148.98	151.10	1.924				
		4	154.21	152.33	151.25	152.59	1.221				
		5	158.94	156.85	160.73	158.84	1.585				
		1	156.10	152.04	1.50.00	156.10	1.400				
	front	1	156.19	153.04	159.09	156.10	1.426				
		2	182.35	180.15	185.5	182.66	1.267				
2.5g		3	194.47	191.66	195.37	193.83	0.912				
		4	198.26	199.43	201.63	199.77	0.806				
		5	199.64	202.33	203.51	201.82	0.935				
	mid	1	105.12	102.2	108.32	105.21	2.499				
		2	143.14	140.34	148.22	143.9	3.261				
		3	157.18	159.65	161.8	159.54	1.887				
		4	164.51	160.1	168.36	164.32	3.374				
		5	165.26	163.44	168.65	165.78	2.158				
	back		83.33	85.12	81.51	83.32	1.4/3				
		2	116.82	119.91	117.11	117.94	1.393				
		3	133.52	135.22	130.35	133.03	2.0181				
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		4	143.07	145.34	146.67	145.02	1.486				
		5	151.31	154.52	156.46	154.09	2.123				
	-										
		1	168.13	165.21	171.32	168.22	1.440				
		2	196.63	199.11	200.57	198.77	0.938				
	front	3	204.54	208.17	206.12	206.27	0.857				
		4	208.57	211.72	213.67	211.32	1.213				
		5	210.03	214.13	217.01	213.72	1.653				
	mid	1	103.22	105.01	100.32	102.85	1.932				
		2	146.43	149.03	143.5	146.32	2.258				
5g		3	164.79	161.6	168.09	164.82	2.649				
		4	172.79	170.87	176.03	173.23	2.129				
		5	175.62	171.83	178.02	175.15	2.548				
		1	49.46	55.02	51.86	52.11	2.276				
		2	79.35	81.15	83.02	81.17	1.498				
	back	3	99.89	102.19	103.02	101.7	1.323				
		4	118.05	116.18	121.01	118.41	1.988				
		5	128.65	125.89	131.25	128.5	2.188				

Table 5.4 Thermocouple measurements (°C) for catalysts 0.5% Pd/Al ₂ O ₃									
Loading	Probe location	LPM	Test-1	Test-2	Test-3	Mean	Error		
		1	125.03	129.4	120.21	124.88	2.166		
		2	144.19	149.133	140.11	144.47	2.1299		
	front	3	146.99	151.902	144.21	147.70	1.835		
		4	146.092	152.232	143.90	147.40	2.035		
		5	145.321	150.11	141.30	145.57	2.078		
			105.05	10004	1.40.51	100.05	1 550		
		1	137.35	136.34	140.51	138.07	1.779		
		2	151.88	147.42	156.61	151.97	3.753		
1g	mid	3	150.18	147.12	153.01	150.11	2.404		
		4	147.85	144.44	151.54	147.95	2.899		
		5	140.77	143.44	145.97	143.39	2.122		
		1	112.12	11100	110 70		0.675		
	back	1	113.43	114.32	119.50	115.75	2.675		
		2	135.67	139.02	142.96	139.22	2.979		
		3	143.59	147.60	146.34	145.84	1.675		
		4	146.34	148.06	145.27	146.56	1.148		
		5	155.05	152.34	153.25	153.55	1.125		
		1	111.26	108.86	109.55	109.89	0.581		
	front	2	125.36	120.77	121.81	122.64	1.134		
		3	133.40	134.13	136.44	134.66	0.747		
		4	116.83	118.24	117.22	117.43	0.344		
		5	100.04	104.45	107.21	103.90	1.704		
2.5g		1	110.07	112.33	113.98	112.13	1.602		
C		2	153.24	156.25	158.43	155.97	2.127		
	mid	3	167.27	166.73	164.01	166.00	1.424		
		4	162.51	166.20	162.41	163.71	1.765		
		5	162.09	165.93	161.18	163.07	2.060		
		1	01.02	02 02	05 72	02 /0	1 755		
	back	2	71.05 117.27	102.00	120.21	120.25	2 2 2 1		
		2	11/.3/	123.08	120.31	120.23	2.331		

							1			
		3	134.12	134.85	136.02	135.00	0.782			
		4	138.24	141.69	142.33	140.75	1.795			
		5	140.53	137.66	141.06	139.75	1.495			
		1	134.32	136.16	135.46	135.31	0.438			
		2	151.06	148.48	146.52	148.69	1.073			
	front	3	140.94	142.91	143.42	142.42	0.617			
		4	137.44	135.43	131.18	134.68	1.507			
		5	121.45	125.67	126.05	124.39	1.203			
	mid	1	101.11	103.08	103.74	102.64	1.117			
		2	137.54	135.78	142.03	138.45	2.631			
5g		3	151.15	152.71	150.34	151.40	0.983			
		4	159.43	156.19	158.10	157.91	1.330			
		5	160.09	158.65	163.57	160.77	2.064			
		1	53.34	52.39	55.04	53.59	1.097			
		2	86.59	92.75	91.65	90.33	2.682			
	back	3	111.28	114.11	117.65	114.35	2.607			
		4	127.30	129.04	125.37	127.24	1.498			
		5	132.44	137.03	135.54	135.00	1.913			

Table 5.5 Thermocouple measurements (°C) for catalysts 5% Pd/Al ₂ O ₃									
Loading	Probe location	LPM	Test-1	Test-2	Test-3	Mean	Error		
		1	161.09	164.12	158.89	161.37	1.237		
		2	183.89	180.53	185.36	183.26	1.167		
	front	3	193.32	195.04	190.77	193.04	1.0128		
		4	197.65	195.28	191.06	194.66	1.574		
		5	199.30	204.32	198.86	200.83	1.429		
		1	148.19	146.45	149.93	148.19	1.420		
		2	169.32	173.24	174.45	172.34	2.189		
1g	mid	3	181.16	179.32	185.04	181.84	2.384		
		4	185.89	188.67	192.36	188.97	2.650		
		5	189.62	191.89	194.04	191.85	1.804		
		1	117.00	114.00	121.20	110.02	2 (14		
	back	1	117.88	114.90	121.30	118.03	2.014		
		2	145.03	142.88	140.56	142.82	1.825		
		3	151.23	150.24	147.77	149.75	1.456		
		4	154.54	157.30	152.83	154.89	1.841		
		5	156.23	159.85	161.32	159.13	2.137		
		1	166.10	155.43	158.69	160.07	2.577		
	front	2	190.53	184.48	187.50	187.50	1.426		
		3	199.35	193.64	196.71	196.57	1.347		
		4	203.23	198.37	205.20	202.27	1.657		
		5	204.82	201.09	202.11	202.67	0.908		
2.59		1	119.83	112.92	118.22	116.99	2.952		
2.08		2	153.15	146.48	149.02	149.55	2.749		
	mid	3	166.54	158.51	163.78	162.94	3.332		
		4	174.55	178.31	176.86	176.57	1.550		
		5	179.54	173.14	178.53	177.07	2.810		
		1	61 21	68.02	66 / 1	66 56	1 971		
	back	2	112.90	110 11	115.60	115.94	1.0/1		
		2	113.80	118.11	113.00	113.84	1./0/		

	3	124.02	127.02	128.15	126.40	1.742
	4	135.74	133.31	136.71	135.25	1.430
	5	144.27	148.54	149.06	147.29	2.145

5.3.2 H₂ conversions

Hydrogen (H₂) conversions for the studied cases are shown in Figure 5.11 for the 0.5% Pt/Al₂O₃ and 1.0% Pt/Al₂O₃ catalysts, respectively. The vertical bar is used in both plots to present the measurement uncertainty. In all cases, H₂ conversion is highest at low flow rates due to the increased residence time and decreases as flow rates increase. For the 0.5% Pt/Al₂O₃ catalyst at a 1 g loading, H₂ conversions are significantly lower compared to the 1.0% Pt/Al₂O₃ catalyst, indicating that the higher platinum content in the 1.0% Pt/Al₂O₃ catalyst has a considerable impact on the catalytic process.

Interestingly, the H₂ conversions for the 0.5% Pt/Al₂O₃ catalyst at 2.5 g and 5.0 g loadings are nearly comparable to the conversion values at 1.0 g and 2.5 g loadings of the 1.0% Pt/Al₂O₃ catalyst, further emphasising the effect of platinum content on catalytic performance. At a 5 g loading of the 1.0% Pt/Al₂O₃ catalyst, H₂ conversions reach nearly 100% across all flow rates, demonstrating the enhanced catalytic efficiency due to the higher platinum content.

Similarly, Figure 5.12 presents the H₂ conversion rates for Pd/Al₂O₃ catalysts at various catalyst loadings and flow rates. As previously discussed, catalyst content plays a crucial role in the catalytic process. Accordingly, the H₂ conversion achieved with 5%Pd/Al₂O₃ is significantly higher than that with 0.5%Pd/Al₂O₃ at the same catalyst loading. Notably, for a catalyst loading of 2.5 g in 5%Pd/Al₂O₃, nearly 100% conversion is observed across all flow rates. However, it is quite convincing that the conversion will be 100% if the catalyst loading is increased, therefore, the catalyst loading of 5 g for in 5%Pd/Al₂O₃ is not tested. The details of the hydrogen measurements for Pt/Al₂O₃ and Pd/Al₂O₃ catalyst pellets at various catalyst loadings are presented in Table 5.6 and Table 5.7, respectively, where the uncertainty calculated is based on the population standard deviation.



Figure 5.11 Effect of catalyst loading on hydrogen conversion for (a) 0.5% Pt/Al_2O_3 (b) 1% Pt/Al_2O_3



Figure 5.12 Effect of catalyst loading on hydrogen conversion for (a) 0.5% Pd/Al_2O_3 (b) 5% Pd/Al_2O_3

Table 5.6 I loadings	Hydrogen co	onversio	n (%) meas	urements fo	or Pt /Al ₂ O ₃	at differen	t catalyst
Catalyst	Loading	LPM	Test-1	Test-2	Test-3	Mean	Error
		1	72.20	74.10	70.97	72.42	0.742
		2	63.96	59.31	60.03	61.10	1.179
	1g	3	55.10	50.45	53.34	52.96	1.106
		4	49.27	43.25	47.30	46.61	1.446
		5	45.13	40.99	43.23	43.12	0.978
		1	92.42	95.63	91.03	93.03	1.926
		2	90.99	93.81	94.22	93.01	1.436
0.5%Pt /	2.5g	3	86.47	89.80	90.12	88.80	1.652
AI_2O_3		4	82.90	83.51	85.05	83.82	0.906
		5	79.12	78.79	80.08	79.33	0.547
		1	06.50	05 10	02.08	05.22	1.071
	5g	1	96.59	95.10	93.98	95.22	1.0/1
		2	95.21	96.12	94.20	95.18	0.782
		3	93.47	95.94	93.19	94.20	1.234
		4	92.98	95.03	92.28	93.43	1.165
		5	90.69	93.78	91.34	91.94	1.331
		1	96.59	94.87	95.34	95.60	0.419
		2	90.18	92.34	89.90	90.81	0.628
	1g	3	85.04	87.62	86.15	86.27	0.611
		4	80.46	83.92	81.39	81.92	0.845
		5	76.50	74.69	77.90	76.36	0.758
I%Pt / Al ₂ O ₃		1	98.12	98.21	98.08	98.14	0.053
		2	97.50	98.15	97.65	97.77	0.279
	2.5g	3	96.18	96.32	97.02	96.50	0.367
	_	4	94.85	95.30	95.88	95.34	0.425
		5	93.50	94.85	94.07	94.14	0.552
		1	I	1	1	I	<u>I</u>

		1	99.39	99.60	99.16	99.38	0.180
		2	99.44	99.30	99.64	99.46	0.141
	5g	3	99.32	99.12	99.18	99.21	0.084
		4	99.55	98.94	98.66	99.05	0.375
		5	99.14	98.51	98.85	98.83	0.254

Table 5.7 I	Hydrogen co	onversio	n (%) meas	urements fo	or Pd /Al ₂ O ₂	3 at differen	t catalyst		
loadings							1		
Catalyst	Loading	LPM	Test-1	Test-2	Test-3	Mean	Error		
		1	86.35	85.56	84.95	85.62	0.331		
		2	81.03	79.93	79.08	80.01	0.461		
	1g	3	77.05	75.72	74.69	75.82	0.557		
		4	72.50	70.90	69.67	71.02	0.668		
		5	69.53	67.75	66.39	67.89	0.741		
		1	I	I	I		I		
		1	91.83	91.02	91.35	91.40	0.332		
0.50/01/		2	90.62	89.69	90.07	90.13	0.381		
0.5%Pd / Al2O3	2.5g	3	87.72	86.50	87.01	87.08	0.499		
1 112 0 5		4	85.62	84.19	84.78	84.86	0.584		
		5	83.44	81.80	82.48	82.57	0.673		
	5g	1	93.73	93.37	92.88	93.33	0.347		
		2	92.71	92.29	91.73	92.24	0.403		
		3	91.65	91.16	90.52	91.11	0.462		
		4	89.73	89.13	88.34	89.06	0.568		
		5	88.33	87.65	86.75	87.58	0.646		
		1	98.20	98.10	97.93	98.08	0.064		
		2	94.19	93.85	93.32	93.79	0.207		
	1g	3	90.08	89.50	88.58	89.38	0.355		
		4	86.50	85.72	84.47	85.56	0.483		
5%Dd/		5	83.80	82.86	81.36	82.68	0.579		
J/0FU/ Al2O2			I	I		I	1		
		1	99.86	99.85	99.55	99.75	0.141		
		2	99.75	99.73	99.35	99.61	0.185		
	2.5g	3	99.55	99.72	99.34	99.54	0.156		
		4	99.24	99.16	99.69	99.36	0.234		
		5	98.87	98.57	98.82	98.76	0.131		

5.4 Conclusions

Experiments on the catalytic combustion of ultra-lean hydrogen/air mixtures (2% H₂) were conducted in a catalytic packed-bed tubular reactor to assess the impact of catalyst loading on the catalytic process. The key findings from the experimental results are summarised below:

- The temperatures at the middle and top locations of the catalyst packed bed increase with rising flow rates for both the Pt/Al₂O₃ and Pd/Al₂O₃ catalysts.
- Super adiabatic temperatures are observed at the bottom location for the 1% Pt/Al₂O₃ catalyst with 2.5 g and 5 g loadings and for the 5% Pd/Al₂O₃ catalyst with 1.0 g and 2.5 g loading.
- For the same catalyst loading with high catalyst content, the overall temperatures are higher compared to the low catalyst content.
- In all cases, H₂ conversion is highest at low flow rates due to the increased residence time, which favours the catalytic process.
- For the same catalyst loading, H₂ conversions are significantly higher for the 1% Pt/Al₂O₃ catalyst than for the 0.5% Pt/Al₂O₃ catalyst, owing to the higher platinum content in the 1% Pt/Al₂O₃ catalyst facilitating the catalytic process. Due to the same reason, the H₂ conversions for 5% Pd/Al₂O₃ are observed higher than 0.5% Pd/Al₂O₃.
- In high content catalyst, the catalytic performance at a low flow rate with a catalyst loading of 1 g is comparable to that observed under other catalyst loadings and flow conditions.

However, the experimental investigation of the catalyst bed reactor revealed two key findings: the catalyst content significantly influences the occurrence of super-adiabatic temperatures, and it determines the minimum catalyst required for catalytic conversion based on operating conditions. These findings are crucial for designing and practical implementation of monolithic reactors with staggered catalyst-coated cylinders, as studied numerically in Chapters 4 and 5.

Chapter 6: Conclusions and future work

The central goal of this work is to explore the intensification of catalytic processes during the combustion process of premixed, lean hydrogen/air mixtures. Both numerical and experimental approaches are employed to analyse the catalytic hydrogen combustion process. Particularly, the following key areas have been investigated:

- Examined the catalytic combustion of premixed lean to ultra-lean hydrogen/air mixtures within a platinum-coated planar burner, achieving zero NO_x emissions.
- Investigated the impact of reactor design on the intensification of catalytic combustion in both planar and non-planar burners.
- Predicted catalytic processes in turbulent reacting flows for both planar and non-planar burners using Large Eddy Simulation (LES).
- Assessed the performance of catalytic combustion in a catalyst-packed bed burner by varying catalyst loading.

6.1 Principal findings

In Chapter 1, this study explores the potential of the integration of hydrogen fuel in the energy sector. Given the challenges of high NO_x emission in hydrogen combustion, the recent research on hydrogen combustion in different applications and their limitations are discussed. Moreover, with a focus on reducing NO_x in hydrogen burners for heating applications, different techniques of recent low NO_x hydrogen burners are discussed. Among these techniques, catalytic-aided hydrogen combustion is the most promising for burning premixed lean hydrogen/air mixtures with near-zero NO_x emissions. Particularly, the choice of this technique is its ability to operate beyond flammability which is not feasible for a non-catalytic approach. However, several research gaps and challenges remain in the development of catalytic burners, and the importance of addressing these issues is thoroughly discussed. In addition, the aims of this work are delineated focusing on exploring simulations and experiments on catalytic burners. The key findings of this work are summarised below.

Additionally, the study focuses on identifying the underlying mechanisms to intensify the catalytic process, thereby reducing the reliance on expensive catalysts.

Chapter 2 examines the premixed combustion of hydrogen/air mixtures in both catalytic and non-catalytic planar reactors, varying the equivalence ratio from 0.10 to 0.20 and the inflow Reynolds number (based on hydraulic diameter) from 114 to 2666. In all cases, the noncatalytic reactor results in incomplete combustion and fails to operate at equivalence ratios below 0.16. In contrast, complete combustion is achieved in the catalytic reactor across all conditions, though the catalyst requirement increases with higher Reynolds numbers. In both catalytic and non-catalytic reactors, flames are observed symmetric about the reactor centreline axis and attached to the reactor walls. Again, the wall temperatures obtained exceed the adiabatic flame temperatures due to the characteristics of hydrogen having a very low Lewis number. However, at low equivalence ratios and Reynolds numbers, combustion is purely catalytic, but as both parameters increase, gas-phase combustion becomes more dominant. The radiation heat loss through the inlet is found significant while operating at a low Reynolds number and responsible for lowering the combustion temperatures. The NO_x emissions in the catalytic reactor are obtained at different flow conditions and controlled by the NO₂ and NNH pathways, with NO_x levels for the lean mixtures remaining below 0.018ppmv.

Chapter 3 investigates the premixed turbulent combustion of lean hydrogen/air mixtures (equivalence ratio of 0.15) in both planar and non-planar reactors. The findings show that reactors with a high surface-to-volume (S/V) ratio significantly enhance the catalytic process, allowing for complete hydrogen combustion over a shorter length of the catalytically coated region. In the non-planar reactor, catalytic surfaces are modified by introducing half and full cylinders to intensify the catalytic process by increasing mass and heat convection. Both Nusselt and Sherwood numbers are calculated over the catalytic surfaces for comparisons among the reactor configurations investigated. The flow and temperature contours from simulations demonstrate that reactor design has a significant impact on the catalytic process. Notably, the intensity of super-adiabatic wall temperatures observed in planar reactors in Chapter 2 is significantly reduced in non-planar reactors with staggered half and full cylinders. In addition, this modified configuration in non-planar results in a 30.7% increase in hydrogen conversion compared to an equivalent planar reactor. Furthermore, discrete catalyst coating in the non-planar reactor proves to be an effective strategy for optimising catalyst usage, reducing the required catalyst by 50% while achieving a catalytic conversion rate of 2 kg/s per unit surface area of the coated region.

Chapter 4 explores the turbulent catalytic combustion of premixed lean hydrogen/air mixtures in both planar and non-planar reactors using Large Eddy Simulation (LES)

modelling. This is the first study to apply LES modelling to the turbulent catalytic process in both reactor configurations. For non-planar reactor, a similar configuration is chosen as studied in Chapter 3 for LES modelling. The LES model utilises a turbulent kinetic energy sub-grid model and the eddy dissipation concept to compute species reaction rates. Extensive validation was performed for both reactor configurations, with experimental results used for comparison. In the planar catalytic reactor, turbulence remains relatively uniform along the reactor length. However, the non-planar reactor, featuring staggered cylindrical rods, significantly increases turbulence, enhancing heat and mass transfer to the catalytic surface and thereby intensifying catalytic combustion. Turbulent flow quantities including the Nusselt, Sherwood and wall shear stress are calculated for comparisons. However, the results reveal that the non-planar configuration is effective in the catalytic combustion process. Unlike the planar reactor, hydrogen conversion in the non-planar reactor is purely catalytic and occurs at lower surface temperatures. The non-planar reactor achieves an H₂ conversion of 80.04% at the reactor exit, compared to 48.1% in the planar reactor. Additionally, the nonplanar reactor achieves equivalent performance to the planar reactor while using 62.5% less catalyst.

Chapter 5 assesses the catalyst's effectiveness in the catalytic hydrogen combustion process by varying catalyst content in support material and the amount of catalyst loading through an experimental investigation. The choice of experimental approach is due to the limitations of addressing the catalyst's effectiveness by numerical methods carried out in Chapters 2 to 4. Again, a catalyst-packed bed tubular reactor is used for experimental analysis. Though this reactor is different in design from the non-planar monolith reactor studied numerically in Chapter 4, both reactors are similar while operating in premixed principle and the catalystcoated cylinders used are of similar size. However, the experimental results reveal that the catalyst content in the cylinder has a significant effect on controlling the super adiabatic temperatures which is found critical in previous chapters. In addition, the minimum use of catalyst loading for effective catalytic hydrogen conversion is observed varying based on the operating conditions. However, these findings are crucial for designing and implementing of the monolith reactors studied in real-world applications.

The concluding remark is that no work in combustion studies can be considered definitive. Every new research builds upon previous studies and, in turn, provides a basis for further investigation. Catalytic hydrogen combustion has been a subject of study for many years, and recently, this technology has gained widespread application. The efficient use of catalysts in combustion processes has become a focal point of interest. This thesis contributes to that interest by presenting new findings rooted in prior research, offering a framework that could guide future investigations.

6.2 Recommendations for the future work

As outlined in Chapter 2, catalytic burners can operate under very lean conditions below the flammability limit, achieving nearly zero NO_x emissions. This makes the concept of premixing hydrogen and air in a gas cylinder below the flammability threshold operationally safe. Moreover, it presents a promising alternative to completely replacing natural gas in cooking and heating applications. Building on this idea, a premixed cylinder containing 2% H_2 in air was used for experimental investigations in Chapter 5. However, further research is needed to evaluate their applicability across diverse operating conditions.

While the catalytic hydrogen combustion under fuel-lean conditions in the reactors studied in Chapter 3 offers benefits in reducing low NO_x emissions, further research is needed to optimise catalyst usage for cost-effective and NO_x -free operations. The non-planar reactor, featuring coated staggered cylindrical surfaces, shows promise in enhancing the catalytic process and could significantly impact the design of catalytic reactors. Future research should explore optimising various design elements, such as reactor geometry, cylinder size and placement, flow distribution improvements, and effective catalyst coating techniques.

Additionally, the numerical investigation of catalytic turbulent combustion using Large Eddy Simulation (LES) in Chapter 4 provides a realistic representation of the catalytic process. However, such studies are limited in the literature when it comes to understanding and predicting the complex catalytic processes associated with varying reactor design parameters. Therefore, it is recommended that future studies utilise advanced LES modelling to enhance the analysis of catalytic combustion processes.

Lastly, exploring different types of catalyst materials is essential, as this area remains underrepresented in catalytic combustion research. Investigating a broader range of catalysts could make catalytic techniques more accessible for a variety of applications

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