# CFD Analysis and Optimization of $CH_4$ Pyrolysis in Commercial Hot Wall Chemical Vapor Deposition Reactor

Thesis submitted in partial fulfillment
for the award of the degree

**Doctor of Philosophy**

*By*

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It is certified that the revised thesis entitled **"CFD Analysis and Optimization of $CH_4$ Pyrolysis in Commercial Hot Wall Chemical Vapor Deposition Reactor"** is being submitted by **Anand Gupta (Roll No. 17041502)** in partial fulfilment for the award of Ph.D in Department of Chemical Engineering IIT(BHU), Varanasi is a record of bonafide work carried out by him.

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# DECLARATION BY THE CANDIDATE

I, Anand Gupta, certify that the work embodied in this thesis is my bonafide work and was carried out by me under the supervision of Dr. Vijay M. Shinde for a period of 7 years from December 2017 to September 2024, at the Department of Chemical Engineering & Technology, Indian Institute of Technology (BHU), Varanasi. The matter embodied in this thesis has not been submitted for the award of any other degree. I declare that I have faithfully acknowledged and given credits to the research workers wherever their works have been cited in my work in this thesis. I further declare that I have not wilfully copied any other's work, paragraphs, text, data, results, etc., reported in journals, books, magazines, reports dissertations, theses, etc., or available on websites and have not included them in this thesis and have not cited as my work.

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It is certified that the above statement made by the student is correct to the best of my Knowledge.

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# Acknowledgement

In the grand tapestry of this doctoral journey, I find myself weaving words of gratitude, and at the forefront stands Dr. Vijay M. Shinde, a mentor par excellence. His guidance has been more than academic; it has been a poetic dance of enlightenment. Like a master potter, he molded my academic clay, quoting the verses of Kabir Das, "गुरु कुम्हार शिष कुंभ है, गढ़ि गढ़ि काढ़ै खोट। अन्तर हाथ सहार दै, बाहर बाहै चोट॥," sculpting not just a researcher but a person of integrity and accountability. To him, I owe not only my academic growth but also the lessons of life. I am indebted to Dr. Shinde not only for his scholarly contributions but also for creating an environment that fostered personal and professional growth. His lessons extended beyond the laboratory, teaching me the art of integrity and the importance of surrounding oneself with a supportive community.

I extend my sincere thanks to Dr. S. S. Rao and Dr. Anurag Kamal for their unwavering mental support, helping me remain resilient through difficult times. Their words of encouragement were a source of strength for me.

Heartfelt thanks to the taxpayers of India, whose contribution forms the foundation of the stipend that supported me during this academic pursuit. The financial support from the Ministry of Human Resource Development (MHRD) added wings to my aspirations, and I acknowledge their role in making this research endeavour possible. The Department of Chemical Engineering & Technology at the Indian Institute of Technology BHU, Varanasi, deserves recognition for providing a nurturing ecosystem for academic excellence. A special note of appreciation goes to my RPEC members, Dr. Jyoti Prasad Chakraborty and Dr. Indrajit Sinha, for their valuable insights and constructive feedback that enriched the quality of this research. To my father, Radhe Shyam Gupta, and my family, who stood as unwavering pillars of love, moral support, and encouragement, I owe immeasurable gratitude.

Lastly, I extend my heartiest thanks to all my well-wishers, whose constant motivation and encouragement were the driving forces that propelled me towards the completion of this thesis.

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**Anand Gupta**

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# Nomenclature

$\vec{u}$ : velocity (m/s)

A: pre-exponential factor in the Arrhenius equation

b: constant

C: hyperparameter

$C_p$: specific heat (kJ/kg)

$D^m$: mass diffusivity ($m^2/s$)

$D^T$: thermal diffusivity ($m^2/s$)

E: activation energy (kcal/mol)

$F_B$: body force (N/kg)

g: acceleration due to gravity ($m/s^2$)

h: enthalpy (kJ/kg)

$h_w$: heat transfer coefficient ($J/m^2sK$)

$h_i^0$ : Standard heat of formation (kJ/kg)

I: unit matrix

$J^h$: energy diffusive flux ($w/m^2$)

J: mass diffusive flux ($kg/m^2s$)

$k_{eff}$: effective thermal conductivity (W/m.K)

MAE: mean square error

M: molecular weight (kg/mol)

Min ($x_n$): minimum value of $x_n$

Max ($x_n$): maximum value of $x_n$

η: outward unit normal

$N_r$: total number of surface reaction

P: pressure (Pa)

$P_0$: initial pressure (Pa)

$x^{mol}$: mole fraction of species

$x_{i,inlet}^{mol}$ : inlet mole fraction of $i^{th}$ species

$x^{wt}$: mass fraction of species

x: independent variable (predictor)

$x^{scaled}$: the scaled value of the data point

$x_B$: best vertex

$x_{c1}$: inside the compression point

Xc2: outside the compression point

$X_G$: good vertex

$x_M$: middle point

$X_W$: worst vertex

$X_R$: reflection point

y: dependent variable (response)

$y_1$: local deposition rate ($kg/m^2s$)

$y_2$: local uniformity index

$y_1^{upper}$ : upper limit deposition rate ($kg/m^2s$)

*Greek symbols*

ρ: density of gas mixture ($kg/m^3$)

τ: stress tensor ($N/m^2$)

μ: gas viscosity (Pa.s)

α: weight for deposition rate

β: weight for deposition rate

$\beta_T$: exponential temperature factor

ω: model coefficient

ε: acceptable margin of error

$\hat{R}$: total production rate of species by chemical reaction (kg/m$^3$.s)

$R^2$: coefficient of regression

$R_i$: molar rate of production of i$^{th}$ species (mol/m$^3$s)

RMSE: root mean square error

s: total area of the substrate (m$^2$)

(s): surface sites

t: time (s)

T: temperature (K)

$T_a$: atmospheric temperature (K)

$T_0$: initial reactor temperature (K)

$T_{ref}$: reference temperature (K)

$T_s$: substrate temperature (K)

$T_w$: reactor wall temperature (K)

$\epsilon$: emissivity factor

$\xi$ : slack variable

$\sigma$: Stefan-Boltzmann constant (W/m$^2$K$^4$)

*Subscripts*

i: i$^{th}$ species

j: j$^{th}$ species

k: k$^{th}$ data point

n: total number of data points

r: r$^{th}$ reaction

*Superscript*

^: estimated value

-: mean value

w: wall surfaces

# Description of non-dimensional numbers

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The influence of various operational parameters on the flow behaviour in the CVD reactor and the film growth performance was studied using multiple dimensionless numbers. The ratio of inertial force to viscous force is given as the Reynolds number (Re):

$$\mathrm{Re} = \frac{\rho u D}{\mu}$$

Where $\rho$ is the density of the gas,

$u$ is the gas flow velocity,

D is the reactor diameter and

$\mu$ is the dynamic viscosity of the gas.

All simulations satisfy the laminar flow condition (Re 2100). The Grashof number (Gr) is defined as the ratio of buoyant force to the viscous force experienced by the fluid particle, which correlates with thermally induced convective fluid heat and mass transport.

$$\mathrm{Gr} = \frac{\rho^2 \mathrm{g} \mathrm{h}^3 (\mathrm{T_S} - \mathrm{T_o})}{\mathrm{T_o} \mu^2}$$

Ts and $T_0$ represent the substrates and the gas mixture's entrance temperature, respectively, while h represents the vertical distance measured from the spinning shaft. Furthermore, the buoyancy-to-inertia ratio, $\mathrm{Gr/Re^2}$, quantifies the relative strength of natural vs forced convection. Natural convection flow would be dominated by a high $\mathrm{Gr/Re^2}$ ratio, resulting in poorer film uniformity due to increased recirculations in the reactor. To obtain a consistent film growth rate, however, the operating conditions and reactor geometry must be carefully chosen. The rotational Reynolds number Rew based on reactor diameter assesses the forced convection effects caused by substrate rotation and is defined as:

$$\mathrm{Re}_\omega = \frac{\rho \omega D^2}{4\mu}$$

w is the rotation of the substrate. The Prandtl number (Pr) is defined as the ratio of momentum to thermal diffusivity.

$$Pr = \frac{C_p \cdot \mu}{k}$$

A low Prandtl number indicates that thermal diffusion has taken precedence over momentum diffusion, resulting in a higher heat transfer rate due to conduction rather than convection. The Peclet number (Pe) is the ratio of heat transfer rate due to convection to heat transfer rate owing to conduction and is stated as:

$$Pe = \mathrm{Pr}.\mathrm{Re} = \frac{\rho u D C_p}{k}$$