**Mixed Convection Heat Transfer of Non-Catalytic CH4 Pyrolysis for Hydrogen Production in a Vertical Tube Using Computational Fluid Dynamics**

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Abstract

Greenhouse gas (GHG) emissions are at the forefront of global concerns. The development of low-carbon hydrogen (H2) production methods has gained prominence due to the ability to substitute fossil fuels in the transport and power generation sectors. Non-oxidative CH4 pyrolysis for low-carbon H2 production holds significant potential to produce H2 and solid carbon without CO2 emissions. A three-dimensional (3D) computational fluid dynamics (CFD) model coupled with chemical reactions and heat transfer was developed to investigate the hydrodynamics, reaction kinetics, and heat transfer of non-catalytic CH4 pyrolysis in a vertical tube reactor. The vertical tube experienced buoyancy forces due to the density variations caused by the significant temperature differences. The mixed convection heat transfer phenomena were observed due to buoyancy forces leading to distortion in the velocity field. The distortion in velocity fields enhanced the heat transfer coefficient in the reactor. The overall convective heat transfer coefficient from wall to fluid was 30.0 W/m2/K. The CFD model is a valuable tool for the identification of chemical reactions and heat transfer coupled with hydrodynamics of the reactor.

**Keywords**: H2 production, non-catalytic CH4 pyrolysis, reactor hydrodynamics, mixed convection heat transfer, computational fluid dynamics (CFD),

**1. Introduction**

Hydrogen (H2), as a clean energy source, offers a promising alternative to fossil fuels and holds the potential to become a leading fuel for sustainable transportation, providing a reliable and secure energy source (Qureshi et al., 2022). The non-oxidative CH4 pyrolysis generates H2 and solid carbons without CO2 emissions, as long as the thermal source remains free from emissions (Catalan and Rezaei, 2020).

The phenomenon occurring inside the noncatalytic CH4 pyrolysis reactor is difficult to investigate due to the high operating temperature (Paxman et al., 2017). Computational fluid dynamics (CFD) simulations are commonly used to study the hydrodynamics and heat transfer characteristics of the reactor following geometric and operational modifications (Ngo and Lim, 2020; Ngo et al., 2023). Ozalp studied the effect of temperature and gas flowrate on methane conversion ($X\_{CH\_{4}}$) for noncatalytic CH4 decomposition (Ozalp and JayaKrishna, 2010). Previously published studies have neglected the effect of buoyancy forces on the heat transfer characteristics in the CH4 pyrolysis reactor for CFD modelling. Mixed convection occurs due to the simultaneous effect of buoyancy forces and externally applied inertia forces and plays a vital role in the reactor heat transfer (Gorai and Das, 2020).

This study aims todevelop a 3D CFD model coupled with chemical reactions, and heat transfer to investigate mixed convection heat transfer for noncatalytic CH4 pyrolysis through a vertical tube. CFD model for CH4 decomposition has the potential to be a useful tool for enhancing heat transfer, particularly in the context of mixed convection caused by buoyancy forces within the reactor.

2. Model description

A single-phase Eulerian 3D CFD model was developed for the noncatalytic CH4 pyrolysis in a vertical tube at 1000 °C and 0.5 LPM. The Eulerian single-phase was modeled by a set of continuity equation, a Navier-Stokes (NS) momentum equation, and an energy equation.$ $The CFD model uses the widely adopted shear-stress transport (SST) k-$ω$ turbulence model for simulating laminar and turbulent mixed flows (Menter, 1994). Figure 1 presents the Eulerian CFD model description of the CH4 pyrolysis reactor.

The 3D geometry and meshing of the computational domain for CH4 pyrolysis are depicted in Figure 2. Poly-hexcore meshes were adopted and mesh independence test was performed to ensure numerical accuracy. The CFD simulation was performed using a commercial CFD code, ANSYS Fluent R2022b (ANSYS Inc., USA) with a 24-core workstation.



Figure 1. Single-phase Eulerian 3D CFD model description



Figure 2. (a) Computational domain (b) meshing of the computational domain

3. Results and Discussions

The reliability and accuracy of the Eulerian CFD model were confirmed through a process of verification and validation. Fig 3 shows the verification and validation of the CFD model. In the verification step, coarse (50,000cells), medium (200,000cells), fine (600,00 cells) meshes were tested for velocity ($u\_{g}$) and temperature ($T$) profiles as shown in Fig 3a. Medium mesh has been selected for the further investigation considering both computational accuracy and cost. Figure 3b shows a comparison of the axial temperature profile in the tube reactor between the CFD results and experimental data. Temperature readings were taken at different axial positions to assess the temperature profile along the tube height (*h*). It can be observed that the CFD simulation results align well with the experimental measurements.

Mixed convection is the combined form of natural and forced convection heat transfer and holds a crucial role in the reactor heat transfer (Gorai and Das, 2020). The impact of buoyancy on heat transfer coefficients can either enhance or impair the process, depending on the flow orientation (upward or downward) (Jackson et al., 1989). This investigation involves the process of downward heating and cooling of fluid in a vertical tube. In Fig 4(a), the velocity ($u\_{g}$) contours are depicted at an operating temperature of 1000 °C and a gas flow rate of 0.5 LPM. The cooling zone of the tube reactor experiences a reversal flow attributed to the buoyancy effect owing to the sudden temperature change (see Fig 4b). Figure 4b shows the temperature contour on the slice normal to the transversal direction (*z=0*). As the gas flows under laminar flow conditions, the gas attains thermal stability (T~ 950 °C) in the pyrolysis zone. Fig 4c shows the CH4 mole fraction ($Y\_{CH\_{4}}$) along the tube reactor. The concentration of CH4​ ($Y\_{CH\_{4}}$​​) decreases as the endothermic reaction progresses, attributed to the high pyrolysis temperature (see Fig 4b). Figure 5a presents a dimensionless parameter, the Richardson number ($Ri=\frac{Gr}{\left(Re\right)^{2}}$) was calculated to investigate the presence of mixed convection phenomena in the CH4 pyrolysis tube reactor. In mixed convection heat transfer, $Ri$ plays a crucial role in assessing the impact of natural and forced convection. When $Ri\gg $1, natural convection dominates the heat transfer process and $Ri\ll 1$. forced convection predominates in governing the heat transfer (Cengel, 2000). At the beginning of the preheat, *Ri* indicates the presence of natural convection, transitioning to forced convection as the gas moves towards the pyrolysis. In the absence of buoyancy forces in the pyrolysis region, the reactor shows a domination of forced convection. In the cooling region, strong buoyancy forces lead to the predominance of natural convection. Fig 5b illustrates the heat transfer coefficient (HTC), calculated along the column height (*h*) using Eq. (1). HTC stabilizes at a constant value (HTC ~ 34 W/m2/K) as the temperature reaches thermal stability in pyrolysis region and fluctuates in the cooling zone due to flow recirculation. The area-averaged overall HTC is 30.03 W/m2/K for the noncatalytic CH4 pyrolysis tube reactor.

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| --- | --- |
| $$HTC=\frac{q\_{W}}{T\_{W}-T\_{c}}$$ | (1) |

where $q\_{W}$ is wall heat flux (W/m2), $T\_{w}$ is wall temperature (°C) and $T\_{c}$ is temperature (°C) at the center of the tube reactor.



Figure 3. Verification of numerical accuracy and validation of the CFD model



Figure 4. (a) Contour of velocity ($u\_{g}$); (b) Contour of temperature ($T$) profile; (c) Axial profile of CH4 mole fraction ($Y\_{CH\_{4}}$)



Figure 5 (a) Axial profile of Richardson number (*Ri*) (b) Axial profile heat transfer coefficient (HTC)

The influence of buoyancy flow becomes more pronounced when operating at lower temperatures, mainly due to the decreased gas velocity. Mixed convection heat transfer phenomena were observed because of buoyancy forces induced by density changes in the tube.

1. Conclusions

The noncatalytic CH4 pyrolysis for low-carbon H2 production in a vertical tube was analysed using computational fluid dynamics. A single-phase Eulerian 3D CFD model coupled with heat transfer, turbulence and reaction kinetics was developed to investigate the presence of mixed convection heat transfer phenomena in the CH4 pyrolysis reactor tube. The findings emphasize that the 3D CFD model has the potential to recognize the buoyancy phenomenon that happens within the reactor during noncatalytic CH4 pyrolysis. The buoyancy effect can be reduced by either reducing temperature differences or increasing the gas flow rate in the reactor.

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References

Catalan, L.J., Rezaei, E., 2020. Coupled hydrodynamic and kinetic model of liquid metal bubble reactor for hydrogen production by noncatalytic thermal decomposition of methane. international journal of hydrogen energy 45, 2486-2503.

Cengel, Y.A., 2000. Heat and mass transfer McGraw-Hill Education New York, NY 10121, United States of America

Gorai, S., Das, S.K., 2020. Studies on Mixed Convection and Its Transition to Turbulence—A Review. 50 Years of CFD in Engineering Sciences: A Commemorative Volume in Memory of D. Brian Spalding, 317-361.

Jackson, J., Cotton, M., Axcell, B., 1989. Studies of mixed convection in vertical tubes. International journal of heat and fluid flow 10, 2-15.

Menter, F.R., 1994. Two-equation eddy-viscosity turbulence models for engineering applications. AIAA journal 32, 1598-1605.

Ngo, S.I., Lim, Y.-I., 2020. Multiscale Eulerian CFD of Chemical Processes: A Review. ChemEngineering 4, 23.

Ngo, S.I., Lim, Y.-I., Kwon, H.M., Lee, U.-D., 2023. Hydrodynamics of molten-metal bubble columns in the near-bubbling field using volume of fluid computational fluid dynamics. Chemical Engineering Journal 454, 140073.

Ozalp, N., JayaKrishna, D., 2010. CFD analysis on the influence of helical carving in a vortex flow solar reactor. international journal of hydrogen energy 35, 6248-6260.

Paxman, D., Trottier, S., Flynn, M., Kostiuk, L., Secanell, M., 2017. Experimental and numerical analysis of a methane thermal decomposition reactor. international journal of hydrogen energy 42, 25166-25184.

Qureshi, F., Yusuf, M., Kamyab, H., Vo, D.-V.N., Chelliapan, S., Joo, S.-W., Vasseghian, Y., 2022. Latest eco-friendly avenues on hydrogen production towards a circular bioeconomy: Currents challenges, innovative insights, and future perspectives. Renewable and Sustainable Energy Reviews 168, 112916.