

Modelling the Effects of Equivalence Ratio on Carbon Nanotubes Diameter in Premixed Flame Based on a Semi-Empirical Approach

Ajani Jimoh Olanrewaju¹, Muhammad Thalhah Zainal^{2*},
Mohd Fairus Mohd Yasin¹, Nurizat Rahman³, Mohammad Hilmi Ibrahim¹

¹High Speed Reacting Flow Laboratory, Faculty of Mechanical Engineering, Universiti Teknologi Malaysia, Johor Bahru, Malaysia

²Takasago Thermal/Environmental Systems Research Laboratory, Department of Mechanical Precision Engineering (MPE), Malaysia-Japan International Institute of Technology (MJIT), Universiti Teknologi Malaysia Kuala Lumpur, Kuala Lumpur, Malaysia

³Energy Markets and Strategy, Energy Systems, DNV Technology Center, 118227 Singapore

ARTICLE INFO

Article history:

Received 27 December 2025

Revised 22 April 2026

Accepted 06 May 2026

Published 15 September 2026

Keywords:

Flame synthesis

Semi-empirical model

Carbon nanotube (CNT)

DOI:

10.24191/jmeche.v23i3.9906

ABSTRACT

In combustion-based synthesis, controlling the morphological properties of carbon nanotubes (CNTs) to meet specific requirements for technological applications remains challenging. This study examines the effect of the equivalence ratio on CNT diameter via flame temperature in a premixed flat flame, using a computational approach validated by experimental data. The overall flame temperature varies from 1 to 2 in increments of 0.2. For each of the six cases, the predicted premixed flat-flame temperature from computational fluid dynamics (CFD) is within 10% of the measured value, demonstrating comprehensive validation. A semi-empirical correlation between CNT diameter and flame temperature is established by using the computed temperature as the dependent variable in an empirical correlation based on existing experimental data. The results show that reducing the equivalence ratio from 2 to 1 increases the average flame temperature, leading to CNT diameter increases of 5% and 20%, respectively. Additionally, the influence of temperature on CNT diameter becomes noticeable only within 6 mm downstream of the flame. Microscopy showed that CNTs with larger diameters (36 – 46 nm) are expected in high-temperature regions ($T > 1300$ K), whereas smaller diameters (29 – 41 nm) are observed in lower-temperature areas ($T < 1300$ K) within the flame.

Introduction

Carbon nanotubes (CNTs) serve as electrode materials, chemical sensors, and catalyst supports in molecular electronics and logic circuits owing to their high aspect ratio, large surface area, and distinctive structural,

^{2*} Corresponding author. E-mail address: mthalhah@utm.my
<https://doi.org/10.24191/jmeche.v23i3.9906>

chemical, and physical properties (Chen et al., 2025; Murjani et al., 2022). Since flames can simultaneously provide the key enablers for CNT growth, namely heat and a carbon source, flame synthesis offers an energy-efficient alternative to the traditional chemical vapour deposition (CVD) process (Zhang et al., 2025; Martin et al., 2021). However, flame synthesis offers limited control over CNT characteristics, such as diameter and alignment, due to the inherently steep temperature gradient within a flame. Premixed and non-premixed flame configurations are commonly used as media for nanomaterial production in combustion-based synthesis technology (Selamat et al., 2023; Zainal et al., 2021). In the former, the hydrocarbon feedstock is combined with air before combustion. In the latter, a carbon source and airflow originate from separate streams.

The separate fuel and oxidizer streams in non-premixed flames enable opposing fuel and air flows, as in counterflow burners. This configuration facilitates the formation of a one-dimensional flat flame with a relatively homogeneous temperature distribution throughout the reaction zone. For example, Heng and co-workers showed that at high strain rates in a 1-D counterflow flame, the temperature field becomes increasingly uniform, revealing a clear window for CNT growth within the flame (Moore, 2022; Heng et al., 2019). In contrast, such a flat flame shape is uncommon in premixed flames. The closest example is the study by Liu et al., who produced a flat flame using two opposed premixed flame-based burners, with premixed flames from each burner impinging on a stagnation plane. However, CNT growth was not investigated, and therefore, no CNT-related analysis was reported (Liu et al., 2017). A previous study configured a premixed flame to impinge on a wall, creating wall stagnation to mimic the stagnation plane typically observed in 1D counterflow non-premixed flames. The study demonstrated the capability to grow aligned CNTs, but the controlling factor, whether in the form of temperature, equivalence ratio, or another parameter, was not examined (Nakazawa et al., 2005).

Recent research shows that the non-premixed flame has a unique combustion parameter, the mixture fraction, which can regulate CNT growth (Cabral et al., 2025; Norizan et al., 2020). It was found that CNTs grow within a specific range of fuel mixture fractions. A similar idea applies to the premixed flame variant through the equivalence ratio (ϕ), which indicates whether the flame operates with excess fuel ($\phi > 1$) or excess air ($\phi < 1$) (Liu et al., 2022). The equivalence ratio serves as a unifying global control parameter that inherently couples the inlet mass flow rates and compositions of both the fuel and oxidizer streams in a premixed flame. This parameter also affects the overall flame temperature, with a premixed flame at ϕ close to unity exhibiting a higher adiabatic flame temperature. Since CNT growth and shape, such as tube diameter, depend largely on the flame's thermal distribution, the equivalence ratio is a vital control parameter in premixed flame synthesis (Zainal et al., 2023).

However, few studies have investigated the equivalence ratio as a parameter for controlling CNT morphology during synthesis. For example, a prior study that produced CNTs on nickel-coated wire mesh within a propane-air premixed flame demonstrated that reducing the equivalence ratio from 2.0 to 1.0 affected the weight and coverage area of the CNTs at the site of growth, where the ratio was highest (Zhang et al., 2020; Daneshvar et al., 2021). Similar findings were observed in another study by Zhang et al. (2022), which examined the morphology and structure of carbon nanotubes produced in a hydrocarbon-rich environment using a premixed propane-air flame with an equivalence ratio of $\phi = 1.8$ –2.2. The results showed that increasing the carbon content in the fuel-rich zone increases the equivalence ratio, thereby expanding the yield area for CNTs. The diameter of CNTs ϕ was also reported to vary. However, more emphasis was placed on microscopy and spectroscopy investigations of the effect of equivalence ratio on CNT growth, rather than on illustrating CNT morphology as a function of both flame temperature and equivalence ratio.

Morphological control of CNTs is primarily studied in chemical vapour deposition (CVD) due to its improved control over growth temperature. For example, a CVD study reported that the growth rate, diameter, and density of synthesized CNTs can be regulated by adjusting the power density during the sputter deposition of the nickel catalyst, thereby altering the catalyst's surface morphology (Lavagna et al., 2021). Generally, it was concluded that nanomaterial size depends on the grain size of the catalyst nanoparticles. Other CVD studies have shown that growth temperature and catalyst nanoparticle size significantly influence CNT diameter (Challiwala et al., 2024; Salah et al., 2021). One study highlighted that active nanoparticle accumulation, which enlarges particles and determines CNT diameter, often results from increased flame temperature, which also frequently causes greater nanoparticle migration across the substrate support surface (Kehl et al., 2023; Hughes et al., 2024). Ibrahim et al. (2023) studied the control of morphology and crystallinity of CNTs in a one-dimensional premixed flat flame using substrate-supported nickel catalysts. The study highlighted that the CNT diameter varies with flame temperature along the height above the burner (HAB).

Results from previous studies on the effects of temperature on catalyst particle sizes indicate that an increase in flame temperature often promotes the migration of nanoparticles on the surface of substrate supports, leading to active agglomeration that increases particle size and, in turn, regulates CNT diameter (Goodman et al., 2021). Since CNT diameter is determined by the size of catalyst particles, which can also be controlled by thermal adjustment, metal particles grow during sintering due to Ostwald ripening or particle migration and coalescence at elevated temperatures, leading to larger particle sizes (Ndayishimiye et al., 2022; Somoju, 2022). Sintering is associated with a loss of reactive surface area due to larger catalyst particles, thereby deactivating a heterogeneous catalyst (Basheer et al., 2020). However, the studies by Moore (2022), Goodman et al. (2021), Meng et al. (2022) and Hamzah et al. (2020) reported that CNT diameter can also be controlled by other parameters, such as the fuel, catalyst type, diluents, oxygen, and species concentration, which play crucial roles in CNT formation and dissociation levels. A study conducted by Salah et al. (2021) showed that varying the temperature alone, whilst other operating conditions were kept constant, significantly affects CNT morphology. The CNT diameter decreases with increasing temperature when H₂ is mixed with CH₄, while the CNT length increases. This observation suggests that carbon atoms play a crucial role in the axial growth of CNTs (Zhang et al., 2020; Li et al., 2020; Sawyer & Hart, 2025).

Hence, the present study, by controlling the flame temperature via the equivalence ratio, explores how this parameter affects CNT diameter within the hydrocarbon-rich environment of a premixed flat flame. Computational Fluid Dynamics (CFD) is used to model the premixed flat flame and to further examine how the equivalence ratio influences CNT diameter, using a newly developed semi-empirical correlation. To the author's knowledge, no previous computational method has been used to investigate a premixed flat flame with a uniform temperature distribution to promote favourable CNT growth. The following section outlines the methodology adopted in the modelling study and concludes that the equivalence ratio is a key control parameter for regulating morphology in flame synthesis.

Methodology

Experimental setup

The CFD results were validated against experimental temperature and flame-image data from Ibrahim et al. (2023). Additionally, the semi-empirical correlation developed in this work was derived by combining CFD results with experimental data on flame temperature and CNT diameter. The outcomes of this computational effort were validated against microscopic photographs of the as-grown CNTs from the experiment. The experimental setup, depicted in Fig. 1, involved a premixed flame fuelled by a mixture of

nitrogen, oxygen, and methane at 27%, 24%, and 49%, respectively. The flow rates used to introduce the gases into the combustion chamber were 0.4, 0.445, and 0.8 standard litres per minute (SLM). The integration of a sintered metal into the main nozzle allowed the premixed fuel–air flow to pass uniformly through the porous medium, resulting in a flat, one-dimensional premixed flame. The experiment utilised a fuel-rich environment with an equivalence ratio of 1.8 to facilitate CNT synthesis, thereby providing an excess of carbon for CNT growth.

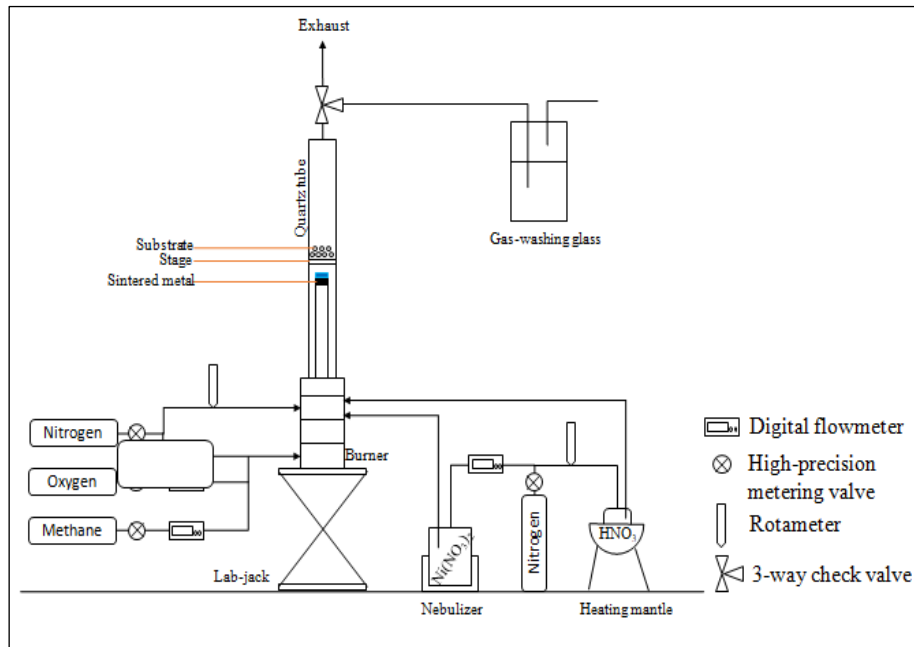


Fig. 1. Experimental setup of the premixed laminar flame synthesis.

As a secondary stream to the primary gas mixture, 6 slpm of nitrogen gas was introduced to prevent carbon precursors from escaping into the atmosphere. In the current investigation, nitrogen gas was also simulated. The CNT growth site was a spherical substrate coated with a nickel nitrate catalyst. A 1.7 MHz mist generator was used to nebulise the nickel nitrate. Nitrogen gas transported the mist, which entered the furnace at 5 slpm and was subsequently deposited onto the spherical substrates. However, the present study does not include catalyst deposition modelling to reduce the computational burden.

Numerical computation

ANSYS Fluent was used to perform computational fluid dynamics (CFD) simulations with the steady-state premixed combustion solver. The distributions of reacting flow velocity, pressure, temperature, and species were obtained by applying the Favre-averaging technique to solve the governing equations for mass, momentum, energy, and species. Equations 1–4 present the governing equations (Posch et al., 2025).

$$\frac{\partial \rho}{\partial t} + \frac{\partial \rho u_i}{\partial x_i} = 0 \quad (1)$$

$$\frac{\partial \rho u_i}{\partial t} + \frac{(\partial \rho u_i u_j)}{\partial x_j} = -\frac{\partial P}{\partial x_i} + \frac{\partial}{\partial x_j} \left[\mu \left(\frac{\partial u_i}{\partial x_j} + \frac{\partial u_j}{\partial x_i} - \frac{2}{3} \delta_{ij} \frac{\partial u_k}{\partial x_k} \right) \right], i = 1, 2, 3 \quad (2)$$

$$\frac{\partial \rho Y_k}{\partial t} + \frac{\partial}{\partial x_i} (\rho (u_i + V_{k,i}) Y_k) = \dot{\omega}_k \quad (3)$$

$$\rho C_p \left(\frac{\partial T}{\partial t} + u_i \frac{\partial T}{\partial x_i} \right) = \dot{\omega}_T + \frac{\partial}{\partial x_i} \left(\lambda \frac{\partial T}{\partial x_i} \right) - \rho \frac{\partial T}{\partial x_i} \left(\sum_{k=1}^N C_{p,k} Y_k V_{k,i} \right) \quad (4)$$

A $k - \omega$ turbulence model was employed to capture local turbulence within the computational domain. The turbulent kinetic energy k and the turbulent dissipation rate ω are defined by Equations 5 and 6, respectively. Variables U and I in Equation 5 denote the mean flow velocity and turbulence intensity, respectively, where a value of 5% is used for the latter variable, accounting for moderate local turbulence levels. Based on Equation 6, C_μ and l are the turbulence model constants taken to be 0.09 and the turbulent length scale, respectively.

$$k = \frac{3}{2} (UI)^2 \quad (5)$$

$$\omega = C_\mu^{\frac{3}{4}} \frac{k^{\frac{1}{2}}}{l} \quad (6)$$

The semi-implicit Method for Pressure-Linked Equations (SIMPLE) scheme was used to handle pressure-velocity coupling, and the second-order upwind scheme was used for grid discretization. The least-squares cell-based (LSCB) approach was used to discretize the gradient terms. A simple kinetic mechanism based on a two-step global reaction for methane-air combustion was employed as the chemical kinetics input, as shown in Equations 7–9. The first reaction represents the oxidation of methane to major combustion products (carbon monoxide and water), as shown in Equation 7. In contrast, the second reaction comprises the forward and reverse reactions of CO₂ formation and dissociation, respectively, as shown in Equations 8 and 9 (Turns, 2012; Gertsyuk, 2023; Picheau et al., 2022).



As shown in Fig. 2, the computational domain of the flat flame is simplified to a two-dimensional, axisymmetric rectangular grid with approximately 38,000 cells. The combustion reactants flow through the domain from the left entrance to the right exit. The bottom boundary represents the flame centerline, which is treated as an axis. Since the primary combustion region is located on the left side of Fig. 2, the grid is denser upstream of the burner.

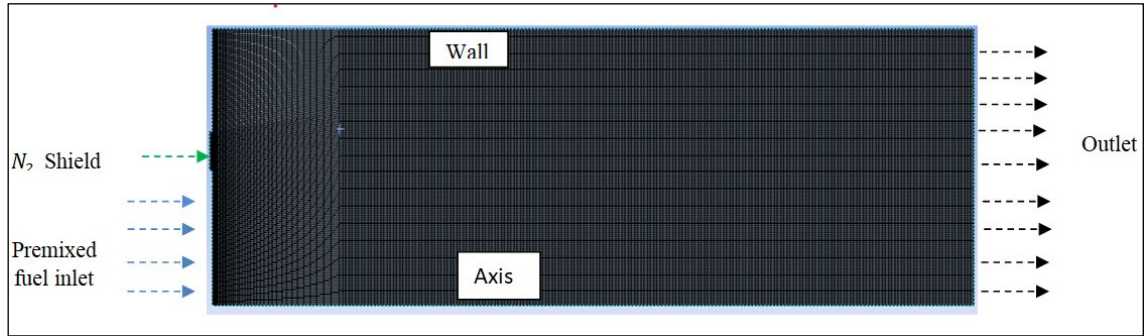


Fig. 2. Schematic of the simplified axisymmetric 2-dimensional computational domain.

First, a baseline case is defined using the experimental flame's operating parameters and an equivalence ratio of 1.8 (Zhang et al., 2022). Equation 10 defines the equivalence ratio (ϕ) (Picheau et al., 2022) as the fuel mass flow rate, $\dot{m}_f$, divided by the air mass flow rate, $\dot{m}_a$, and by the stoichiometry. As indicated in Table 1, the equivalence ratio varies from stoichiometric conditions of 1.0 to fuel-rich conditions of 1.2, 1.4, 1.6 and 2.0, creating six cases. 1.4, 1.6 and 2.0, creating six cases.

$$\phi = \frac{\dot{m}_f / \dot{m}_a}{(\dot{m}_f / \dot{m}_a)_{stoic}} = \frac{\dot{m}_f / \dot{m}_a}{s_a} \quad (10)$$

Table 1. Variation of the equivalence ratio and the corresponding mass flow rate of the combustion reactants used in the present study

Case	Equivalence ratio (ϕ)	CH ₄ mass flow rate (slpm)	O ₂ mass flow rate (slpm)	Total mass flow rate (CH ₄ +O ₂) (kg/sec)	Minimum N ₂ gas flow rate (slpm)	Premixed fuel inlet mass flow rate (kg/s)
1	1	0.4	0.8	2.22×10^{-5}	3	04×10^{-5}
2	1.2	0.4	0.67	1.93×10^{-5}	2.5	78×10^{-5}
3	1.4	0.4	0.57	1.71×10^{-5}	2	5.59×10^{-5}
4	1.6	0.4	0.5	1.15×10^{-5}	1.5	4.46×10^{-5}
5	1.8 (baseline)	0.4	0.445	1.43×10^{-5}	0.8	2.98×10^{-5}
6	2.0	0.4	0.4	1.33×10^{-5}	0.3	1.91×10^{-5}

Due to less-than-ideal growth conditions, fuel-lean conditions with an equivalence ratio ϕ less than unity are not considered. To adjust the equivalence ratio in the present experiment, the inlet air mass flow rate is varied while the fuel flow rate is kept constant at 0.4 slpm. An overview of the boundary conditions applied to the CFD flame model is shown in Table 2. Note that the specified boundary conditions cover the baseline case equivalence ratio, ϕ , of 1.8.

Except for the wall, which is subject to heat flux boundary condition, all other boundaries are set to the room temperature of 300 K. For the burner inlet and shield gas boundaries, intake mass flow rates of 2.98×10^{-5} and 1.21×10^{-5} respectively, are specified. A no-slip condition is assumed at the wall boundary. Based on Table 2, the mass fractions of the reactants CH₄, O₂, and N₂ are represented by Y_{CH_4} , Y_{O_2} , and Y_{N_2} , respectively. The computational time for a single case was approximately 15 minutes on a machine with an Intel Core i7-8700 CPU at 3.20 GHz, using all four cores.

Table 2. Summary of boundary conditions for premixed flat flame (baseline model)

Parameter	Burner inlet	N ₂ Shield gas	Wall	Outlet
T (K)	300	300	Heat flux	300
P (Pa)	--	--	--	Pressure outlet
$\dot{m}$ (kg/s)	2.98×10^{-5}	1.21×10^{-5}	No-slip	--
Y_{CH_4}	0.24	0	--	--
Y_{O_2}	0.27	0	--	--
Y_{N_2}	0.49	1	--	--

Development of semi-empirical correlation

Experimental data for CNT diameter is plotted against the measured CNT growth temperature. These experimental measurements were obtained from a previous flame synthesis experiment (Ibrahim et al., 2023) conducted using the same premixed flame configuration and operating conditions as shown in Fig. 3. When extracting the data and using them to develop correlations, the HAB is implicitly accounted for through its associated temperature and is not treated as an independent variable in the correlation. Data fitting for the $d = f(T)$ relationship is carried out, where linear, exponential, polynomial, and power models were assessed. The 2nd order polynomial resulted in a reasonable fit with R^2 value of 0.999, as shown by Equation 11. It is noted that in Equation 11, the units for the CNT diameter (d) and the average growth temperature (T) are nanometers and Kelvin, respectively.

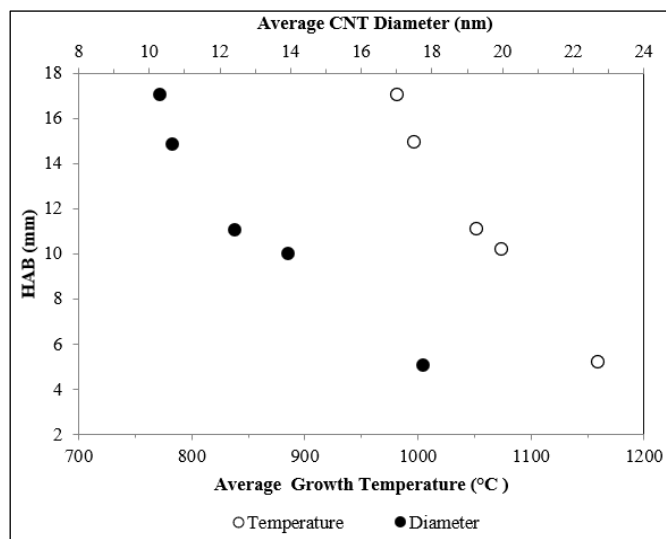


Fig. 3. Average CNT diameter distribution with relative temperature for premixed flame.

$$d = 1.34 \times 10^{-5} T^2 - 0.24 T + 120 \quad (11)$$

The predicted flame temperature for each of the six cases was determined using CFD simulation. The semi-empirical correlation in Equation 11 used the average temperature 5 to 15 mm above the burner (HAB) as the input temperature for the baseline case, and the corresponding diameter was computed. The semi-empirical model relates the predicted flame temperature to the CNT diameter. The process was repeated for the six samples with diameters of 1.0, 1.2, 1.4, 1.6, and 2.0.

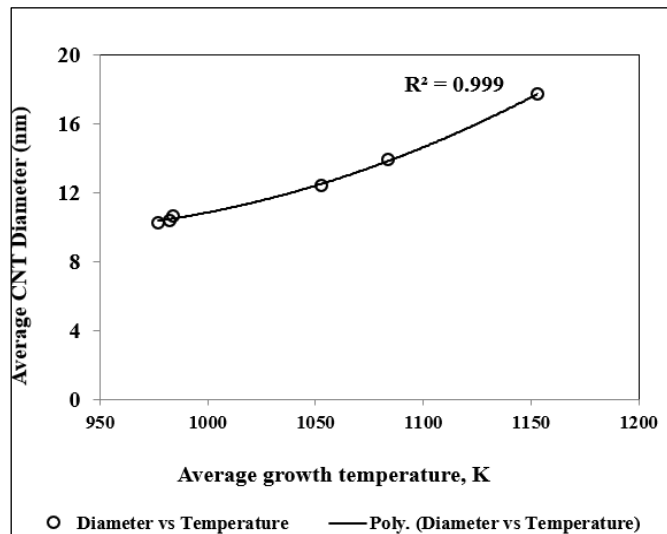


Fig. 4. Scatter plot for experimentally determined CNT diameters at corresponding growth temperatures together with the fitted polynomial model.

Fig. 4 shows the polynomial fit to the empirical relationship between the average CNT diameter and the average CNT growth temperature. This diameter–temperature correlation is used as a post-processing model, in which CFD temperature results are substituted into the correlation to yield predicted CNT diameters. The input temperature corresponds to the average temperature for a given HAB. Thus, HAB is indirectly incorporated into Equation 11. For the present study, HABs ranging from 5 to 17 mm in 2 mm increments are considered. By applying CFD data to the empirical relationship, the semi-empirical model relating CNT diameter to growth temperature is established. The modelling process was then repeated for six operating conditions with equivalence ratios of 1.0, 1.2, 1.4, 1.6, and 2.0. The present semi-empirical approach is reproducible for other flame configurations; direct application to other flame systems would require recalibration using the corresponding experimental data for flame temperature and CNT diameter.

Results and Discussion

Model validation for baseline case

Validation of the flame shape and temperature for the baseline case with $\phi = 1.8$ was carried out. Before the parametric study of the effects of equivalence ratio on CNT diameters, the shape and size of the predicted thin reaction zone, where heat is released due to the combustion of a methane-air mixture, are compared with those of the experimental image, as shown in Fig. 5(a). The line-of-sight images of the predicted and experimental flame fronts are highly similar. The heat-release contour matches the regime well, with a faint bluish flame colour indicating the maximum temperature in the experiment. Fig. 5(b) compares the predicted temperature distribution in the right panel with the experimental flame image. The region of high temperature in the upstream regime, close to the burner inlet, and the gradual decrease towards the downstream location, as predicted by CFD, compare well with the image of the premixed flame, which exhibits an intense bluish-white glow near the inlet and a faded colour at greater axial distances. Overall, the CFD flame model effectively simulates the physics of the flame.

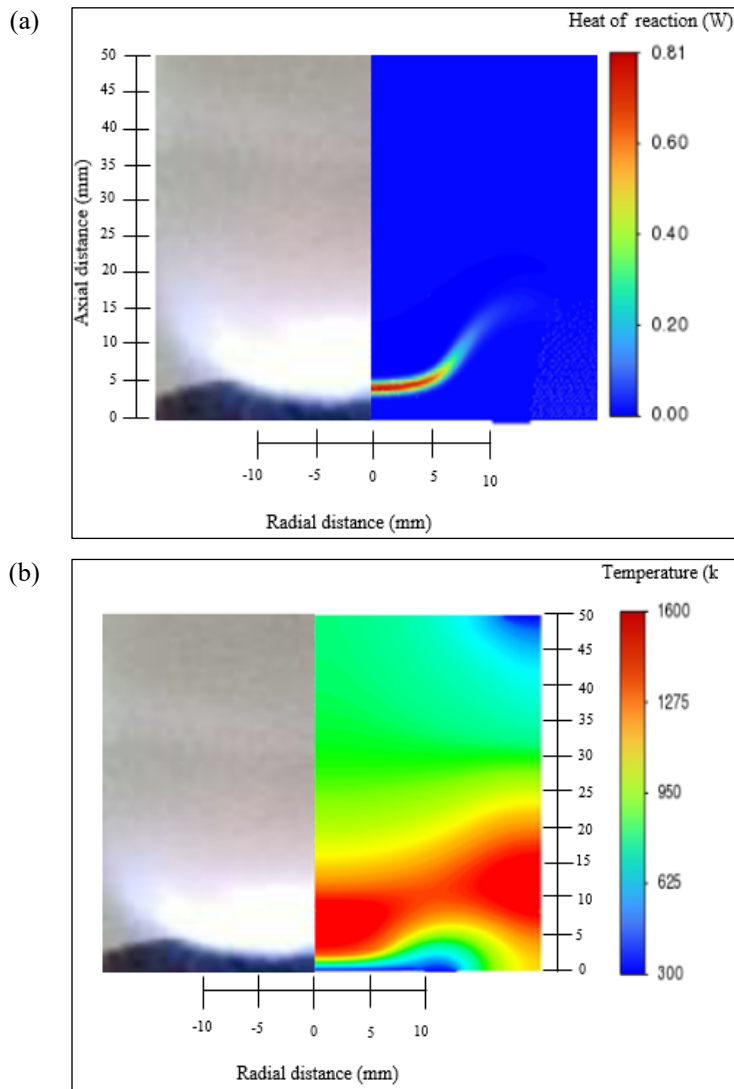


Fig. 5. Validation of the (a) predicted flame front via heat release contour (right panel) against that of the experimental flame (left panel) and (b) flame temperature contour for the baseline case with $\phi = 1.8$.

Fig. 6 illustrates the radial growth temperature at HABs of 5, 7, and 9 mm. The solid lines show the predicted temperatures, while the discrete points show the measured temperatures from the actual experiment. The temperature is calculated along the centreline ($R = 0$) to a radial distance of 8 mm for all cases. The CFD flame model reproduces the decrease in the actual flame temperature with increasing height above the burner, due to the greater distance from the hot flame front. Additionally, the model reasonably predicts a downward slope as radial distance increases. This temperature gradient from the flame centreline towards the far field can be attributed to cold shield gas in the flame's outer annulus (Ibrahim et al., 2023).

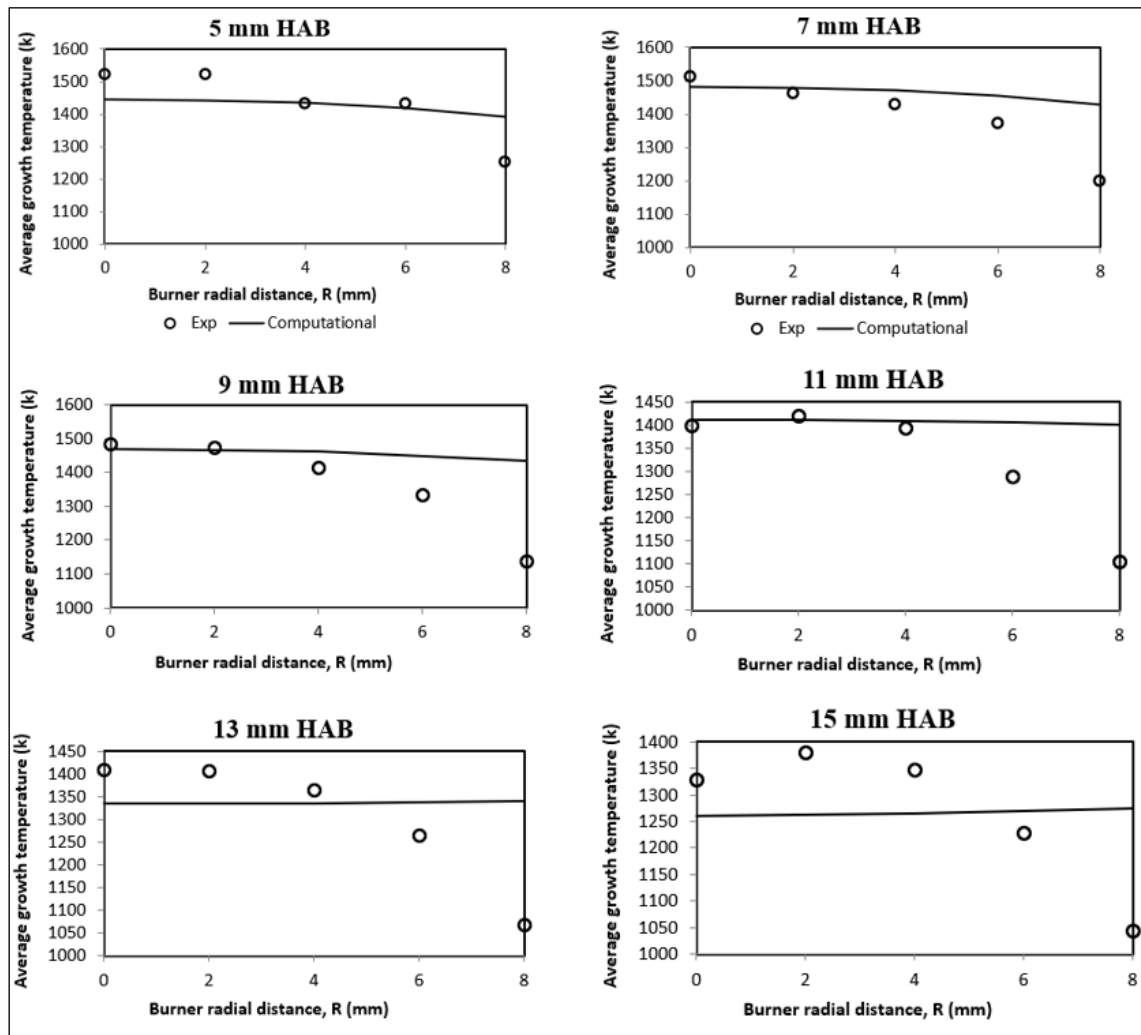


Fig. 6. Validation of radial flame temperature at various burner (HAB) heights at the baseline case of $\phi = 1.8$.

Regarding validation accuracy, the average percentage error between the actual and predicted temperature values is less than 10% in each case, except at 5 mm HAB, where the error is higher due to overprediction of the experimental data in the far field. At 5–11 mm HAB, close to the burner centreline, the flame model underpredicts the temperature profile by well under 5%. A more noticeable underprediction, less than 10%, occurs at higher burner heights, from 13 mm HAB to 15 mm HAB near the burner centreline.

Conversely, an overprediction of the temperature of 5–25% occurs at the radial distance $R = 8$ mm across all HAB. Such a compromise in accuracy is expected, as previous studies have reported a 15–25% discrepancy in temperature prediction between a simulation based on simple chemical kinetics and a detailed one (Zainal et al., 2023). The temperature overprediction observed at cap $R = 8$ mm across all HAB heights is primarily attributed to the use of a simplified two-step global chemistry mechanism, which reproduces overall heat release and flame structure but exhibits reduced accuracy in regions with strong radial temperature gradients. Importantly, CNT growth for all validated cases shown in Fig. 6 occurs in

regions close to the centreline, within a radial distance of 4–6 mm [23]. Since the present CNT diameter analysis is based exclusively on these experimentally validated growth regions, the localized discrepancy in the far-field region is assumed to have a negligible impact on the conclusions of this study. Notwithstanding, the flame model captures the temperature profile throughout the flame with sufficient accuracy despite the simple chemistry approach used in the present study. Fig. 7 shows the predicted axial temperature profile along the flame centreline with $R = 0$ mm. The highest-temperature region lies between 7 and 9 mm HAB in the axial direction, indicating the hot reaction zone. The predicted temperature is notably higher at upstream locations near the burner inlet. These observations align with the actual flame stabilising the burner upstream at 7 mm HAB.

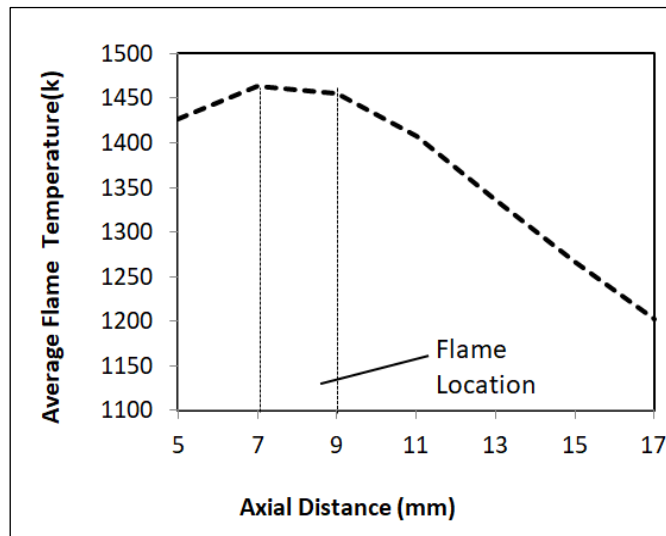


Fig. 7. Numerical results of the axial temperature profile at $R = 0$ with the predicted flame location.

Effects of equivalence ratio on CNT diameter

The effect of the equivalence ratio on CNT diameter is examined over the range of ϕ described in Table 1. Since the equivalence ratio directly influences the flame temperature and, consequently, CNT diameter, an overview of the temperature distribution within the flame is considered necessary, as shown in Fig. 8. For conciseness, only the temperature distribution near the reaction zone at 7 and 9 mm HAB, as well as at upstream (5 mm HAB) and downstream (15 mm HAB) positions relative to the reaction zone, is presented. Fig. 8 shows the predicted temperature distribution at varying HAB and equivalence ratios. The high-temperature range of 1400–1550 K at 7 and 9 mm HAB corresponds to the flame location, as shown earlier in Fig. 7. The case of $\phi = 1$ consistently shows the highest temperature across all HAB. This is expected, as the maximum heat release occurs at the stoichiometric proportion of fuel and air, where the equivalence ratio (ϕ) is unity; away from this value ($\phi < 1$ or $\phi > 1$), the flame temperature will be lower (Sawyer & Hart, 2025). As the equivalence ratio decreases, the radial temperature also decreases.

The temperature range is significantly lower, between approximately 1300 and 1400 K, and further from the reaction zone at 5- or 15-mm HAB. Away from stoichiometric conditions, at higher equivalence ratios of 1.2, 1.4, 1.6, 1.8, and 2.0, the temperature distribution decreases consistently. Fig. 7 provides the basis for determining the average temperature, which is used to plot the semi-empirical relationship between flame temperature and CNT diameter in Fig. 8. Based on Fig. 8, a triaxial plot is presented with equivalence ratio as the independent variable and flame temperature and CNT diameter as the dependent

variables to systematically analyze the three parameters. Note that the range of the equivalence ratio under study is limited to 2.0. Under ultra-rich conditions (> 2.2), growth competition with amorphous carbon can become significant at higher carbon contents (Muchuweni et al., 2022; Zainal et al., 2020). Such growth physics are not the primary focus of the present study.

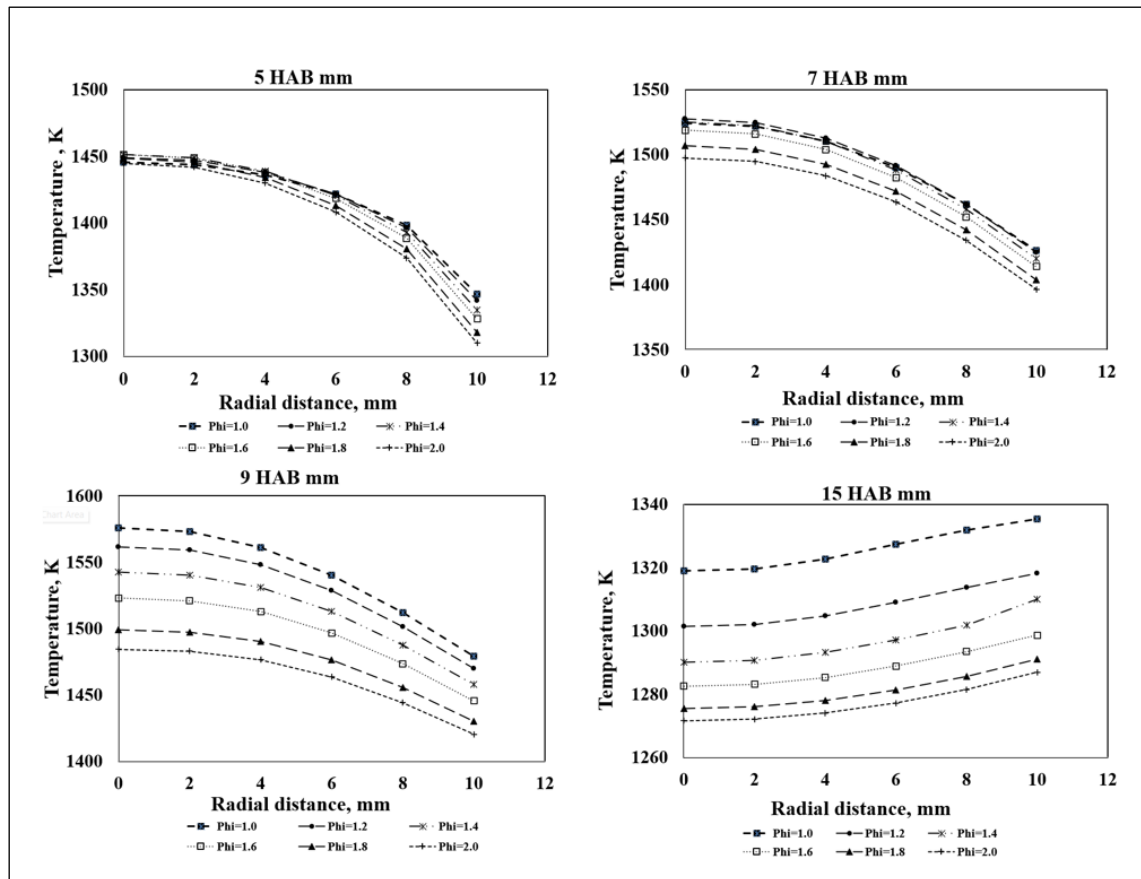


Fig. 8. Temperature profile with varying equivalence ratios at 5, 7, 9, and 15 mm HAB.

Morphological control of CNTs is often explored in chemical vapour deposition (CVD) because of its ability to precisely control growth temperature. For instance, CVD studies have shown that adjusting power density during nickel catalyst sputter deposition can modify the catalyst's surface morphology, thereby regulating the growth rate, diameter, and density of the resulting CNTs (Lavagna et al., 2021). Typically, the size of the nanomaterial is tied to the grain size of the catalyst nanoparticles. CVD research has also highlighted that both growth temperature and catalyst nanoparticle size play a significant role in determining CNT diameter (Challiwala et al., 2024; Salah et al., 2021). Research has shown that higher flame temperatures can increase nanoparticle migration onto the substrate support, causing particles to accumulate and grow larger, which in turn influences CNT diameter (Kehl et al., 2023). Ibrahim et al. (2023) explored the control of CNT morphology and crystallinity using nickel catalysts in a premixed flat flame. They found that the CNT diameter changed with flame temperature at different heights above the burner (HAB). The equivalence ratio (Φ) in premixed flames indicates fuel-rich ($\Phi > 1$) or fuel-lean ($\Phi < 1$) conditions (Heng et al., 2019). Φ affects flame temperature, which peaks near unity and, in turn, influences

CNT growth and morphology, making it a key control parameter in premixed flame synthesis (Zainal et al., 2023). Research on the use of the equivalence ratio to control CNT morphology is limited. One study using a propane-air premixed flame with nickel-coated wire mesh found that lowering the equivalence ratio from 2.0 to 1.0 affected CNT weight and coverage area, particularly at the highest ratios (Zhang et al., 2020; Daneshvar et al., 2021).

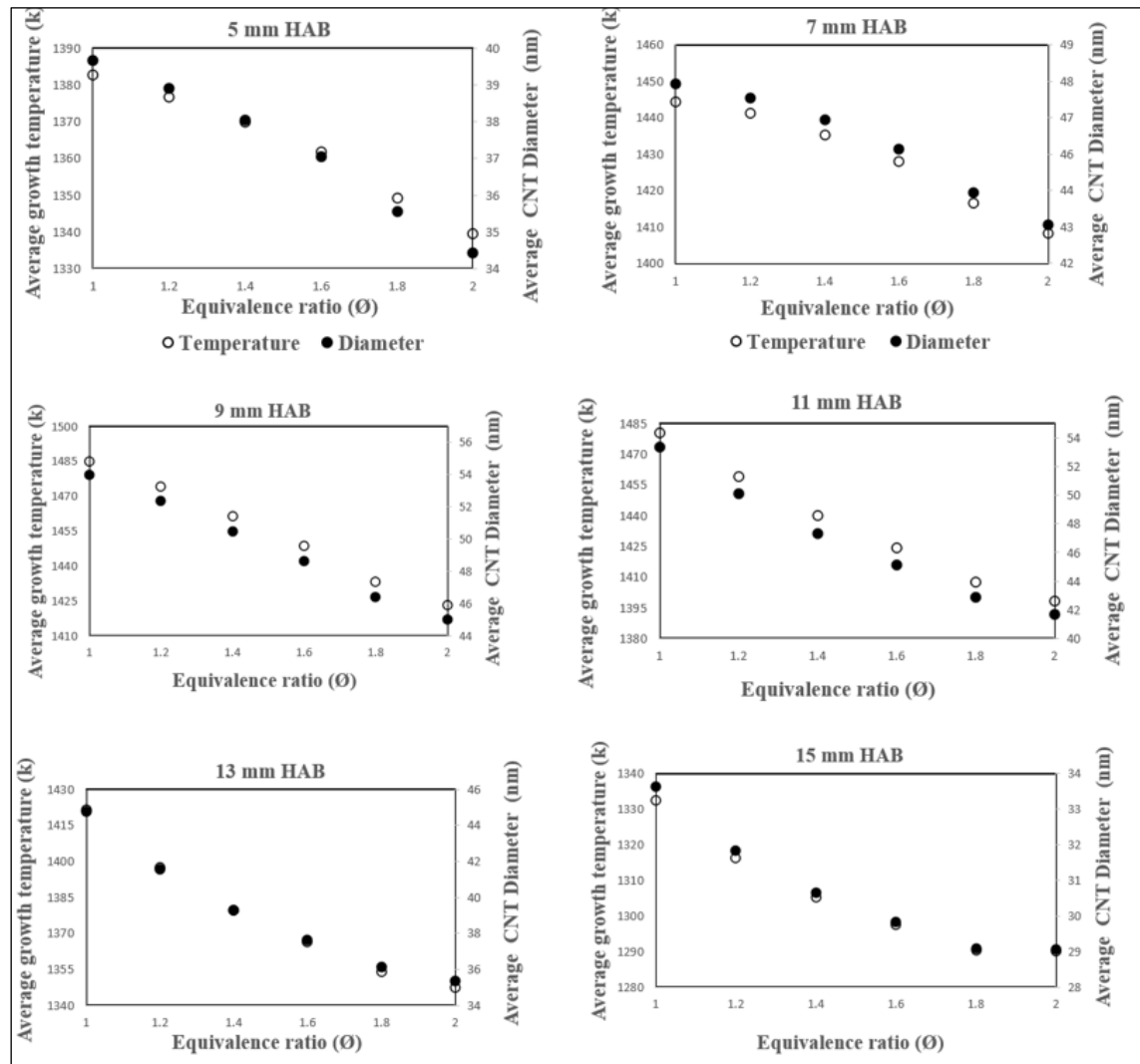


Fig. 9. Equivalence ratio variation with flame temperature and CNT diameter at 5, 7, 9, 11, 13, and 15 mm HAB.

As shown in Fig. 9, the flame temperature increases as the equivalence ratio shifts from rich to stoichiometric conditions, as expected, since complete combustion, which produces the highest heat release, is more prevalent under stoichiometric conditions. Interestingly, the CNT diameter also increases as the equivalence ratio decreases, as observed consistently across all HABs. Across all triaxial plots in Fig. 8, the predicted CNT diameter at HABs of 5, 7, 9, 11, 13, and 15 mm ranges between 29 and 55 nm. The overlap between the predicted average temperature (white dots) and the diameter (black dots) indicates that

the equivalence ratio affects the local temperature and, consequently, the CNT diameter, as predicted by the correlation in Equation 11. The diameter is highly sensitive to temperature, with a 5% variation in average temperature across HABs resulting in approximately a 20–30% change in CNT diameter. While the variation in diameter is noticeable at varying HAB, it must be noted that HAB is not the primary factor that influences CNT growth characteristics. Rather, it reflects the spatial manifestation of the temperature field established by a given equivalence ratio.

In contrast to CVD studies demonstrating that CNT diameters are related to catalyst particle size (Goodman et al., 2021; Ndayishimiye et al., 2022), the present study suggests that diameter depends on the local flame temperature, as indicated by the developed diameter–temperature correlation, which is obtained by adjusting the flame's equivalence ratio. This observation that the equivalence ratio is the primary influence on CNT growth aligns with studies in the open literature (Zhang et al., 2020; Daneshvar et al., 2021). For instance, a previous flame synthesis experiment that employed a propane- air premixed flame with a nickel- coated wire mesh found that increasing the equivalence ratio from 1 to 2 substantially affected the weight and coverage area of CNTs, with the most pronounced growth observed at the location with the highest equivalence ratio (Zhang et al., 2020). Complementary findings were reported by Zhang et al. (2022), who investigated the morphology and structure of CNTs produced in a hydrocarbon- rich environment using a propane- air premixed flame with an equivalence ratio range of $\phi = 1.1$ – 2.2 . The results suggest that optimizing the equivalence ratio can significantly enhance CNT yield and quality. It can be inferred from Fig. 9 that larger catalyst particles are more likely to form in flames with an equivalence ratio closer to 1 within the fuel- rich regime. The formation of larger catalyst particles at higher temperatures, as the equivalence ratio nears unity, can be attributed to Ostwald ripening and particle migration and coalescence (PMC), which are more pronounced at elevated temperatures (Goodman et al., 2021; Meng et al., 2022). As carbon content increases at higher equivalence ratios, the average diameter of the CNTs decreases. On the other hand, the absence of a radial (diameter- wise) contribution from carbon to CNT growth indicates that the carbon is either consumed for growth along the axial direction (length- wise) or transformed into other combustion by- products, including amorphous carbon and polycyclic aromatic hydrocarbons (PAHs). This notion of competing effects between CNTs and carbon- based particulates, such as PAHs, amorphous carbon, and soot, has been suggested by previous studies (Xu et al., 2006; Wang et al., 2011). While such observations are reported in experimental studies, further investigation is required to confirm them within a modelling- based framework.

As shown in Fig. 9, reducing the equivalence ratio from 2 to 1 affects CNT diameter only within a limited downstream range of the flame, between 9 and 13 mm HAB (with the flame located at 7 mm HAB), where CNT diameter increases by 20–30% within this HAB span. Conversely, at positions below 9 mm HAB and beyond 15 mm HAB, a similar reduction in the equivalence ratio results in only up to a 10% increase in CNT diameter. This observation is attributed to the local temperature distribution, as shown in Fig. 8. HABs below the reaction zone at 7 mm HAB exhibit low temperatures of 1300–1400 K. On the other hand, the temperature at 9 mm HAB is much higher, ranging from 1400 to 1500 K. The effect of the equivalence ratio on local flame temperature at this HAB is also more pronounced, as indicated by the relatively sparse spacing of the temperature plots. However, at 15 mm HAB, the diameter shown in Fig. 9 does not vary notably despite the sparse spacing observed at that HAB, as shown in Fig. 8, implying adverse effects of low temperature on CNT growth.

The findings from the present computational studies are further supported by FESEM images of the as- produced CNTs at HABs of 5, 10, and 15 mm for the baseline case, as shown in Fig. 10. Based on these images, CNT diameter decreases with increasing HAB at lower temperatures. A wider diameter distribution, ranging from 46 to 50 nm, occurs at lower HAB between 5 and 10 mm in the high- temperature region ($T > 1300$ K). Conversely, a narrower diameter distribution, between 20 and 35 nm, appears in the low- temperature region ($T < 1300$ K) at a higher HAB of 11–15 mm. This observation of the diameter

distribution aligns with the predicted CNT diameters of 36–46 nm at 5–9 mm HAB and 29–41 nm at 11–15 mm HAB for the baseline case, as shown in Fig. 10.

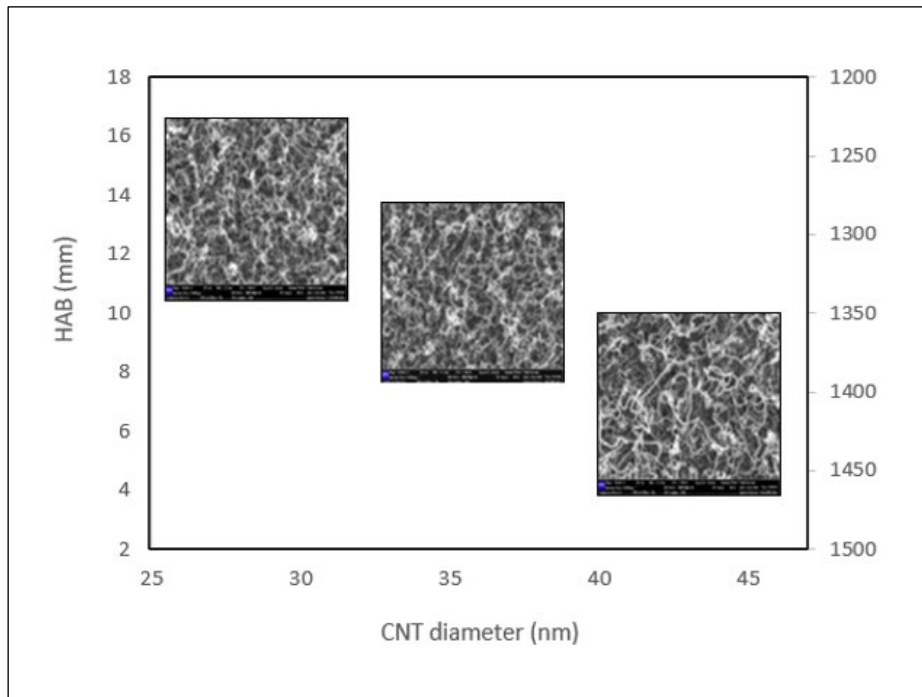


Fig. 10. Field Emission Scanning Electron Microscopy (FESEM) images at 50,000 $\times$ magnification of 5, 10, and 15 mm HAB for the baseline equivalence, $\phi = 1.8$.

Conclusion

A parametric study of the effect of temperature on CNT diameter is conducted as a function of the equivalence ratio. The study is set in a premixed flat flame. Validation results for the baseline case, with the equivalence ratio set to 1.8, showed a satisfactory agreement between CFD predictions of flame temperature and experimental values, with errors well within 10%. The distribution of CNT diameter at temperatures of 1000, 1200, 1400, 1600 and 1800 K for equivalence ratios of 1.0, 1.2, 1.4, 1.6 and 2.0 was determined using a newly developed semi-empirical correlation between temperature and CNT diameter, based on published experimental results. Computational results showed an increase in CNT diameter as the equivalence ratio decreased, a trend consistently observed across all flame heights studied. This suggests that the equivalence ratio is a potential control parameter in premixed flames. Further comparison of the computational results with actual microscopy images of CNT diameter confirms that CNTs with smaller diameters are observed in lower-temperature regions below 1300 K. In contrast, CNTs with larger diameters are observed at higher temperatures above 1300 K. The results also indicate that, for a premixed flat flame, the effect of the equivalence ratio on CNT diameter is significant only within a limited downstream location of the flame.

Acknowledgements/ Funding

Special thanks to the team at the High-Speed Reacting Flow (HiREF) Laboratory for their invaluable contributions and support in making this work a reality. The research was funded by Universiti Teknologi Malaysia (UTM) through the UTM Fundamental Research (UTMFR: PY/2024/01611) grant with cost centre number Q.J130000.3824.23H94. This research was also supported by Universiti Teknologi Malaysia (UTM) through the Prototype Development Fund (PDF Cycle 2: PY/2024/00430) grant with cost centre number Q.J130000.4424.4J676.

Conflict of Interest Statement

On behalf of all authors, the corresponding author states that there is no conflict of interest.

Authors' Contributions

The authors confirm their contribution as follows: study conception and design: A.J. Olanrewaju, M.T. Zainal, and M.F. Mohd Yasin; data collection: A.J. Olanrewaju, N. Rahman, and M.H. Ibrahim; analysis and interpretation of results: A.J. Olanrewaju and M.T. Zainal; draft manuscript preparation: A.J. Olanrewaju, M.T. Zainal. All authors reviewed the results and approved the final version of the manuscript.

Declaration of Generative AI in the Writing Process

During the preparation of this work, the authors used COPILOT and META AI to paraphrase and proofread. After using this tool/service, the authors reviewed and edited the content as needed and takes full responsibility for the content of the publication.

Data Availability/ Supplementary Materials

Data and materials are available on request.

Ethics Statement

The authors declare that this research did not involve human or animal subjects. All experimental procedures were performed following the institutional Safety, Health, and Environmental (HSE) protocols of Universiti Teknologi Malaysia (UTM).

Reference

Basheer, H. J., Baba, K., & Bahlawane, N. (2020). Thermal chemical vapor deposition of superblack randomly oriented carbon nanotube coatings. *Physica Status Solidi (A)*, 217(8), 1900704. <https://doi.org/10.1002/pssa.201900704>

<https://doi.org/10.24191/jmeche.v23i3.9906>

- Cabral, T. O., Amama, P. B., & Pourkargar, D. B. (2025). Multiscale-multiphysics predictive modelling of chemical vapour deposition processes for carbon nanotube synthesis. *Chemical Engineering Science*, 305, 121137. <https://doi.org/10.1016/j.ces.2024.121137>
- Challiwala, M. S., Ibrahim, G., Choudhury, H. A., & Elbashir, N. O. (2024). Scaling up the advanced dry reforming of methane (DRM) reactor system for multi-walled carbon nanotubes and syngas production: an experimental and modelling study. *Chemical Engineering and Processing: Process Intensification*, 197, 109693. <https://doi.org/10.1016/j.cep.2024.109693>
- Chen, Y., Zhang, Z., Chen, J., Shen, P., Shi, Z., & Zhang, G. (2025). Carbon nanotubes: from disordered ensembles to van der Waals crystals. *The Innovation*, 7(3), 101017. <https://doi.org/10.1016/j.xinn.2025.101017>
- Daneshvar, F., Chen, H., Noh, K., & Sue, H. J. (2021). Critical challenges and advances in the carbon nanotube–metal interface for next-generation electronics. *Nanoscale Advances*, 3(4), 942–962. <https://doi.org/10.1039/d0na00822b>
- Gertsuik, M. (2023). Prediction of pollution level between measurement points by mathematical modeling using interpolation and recursion. *Science and Innovation*, 19(3), 65–70. <https://doi.org/10.15407/scine19.03.065>
- Goodman, E. D., Carlson, E. Z., Dietze, E. M., Tahsini, N., Johnson, A., Aitbekova, A., Taylor, T. N., Plessow, P. N., & Cargnello, M. (2021). Size-controlled nanocrystals reveal the spatial dependence and severity of nanoparticle coalescence and Ostwald ripening in sintering. *Nanoscale*, 13(2), 930–938. <https://doi.org/10.1039/d0nr07960j>
- Hamzah, N., Mohd Yasin, M. F., Mohd Yusop, M. Z., Saat, A., & Mohd Subha, N. A. (2020). Growth region characterization of carbon nanotubes synthesis in heterogeneous flame environment with wire-based macro-image analysis. *Diamond and Related Materials*, 99, 107500. <https://doi.org/10.1016/j.diamond.2019.107500>
- Heng, E. W., Zainal, M. T., Mohd Yasin, M. F., & Hamzah, N. (2019). Effects of flame structure and flame strain on the growth region of carbon nanotubes in counter-flow diffusion flame. *CFD Letters*, 11(6), 72–85.
- Hughes, K. J., Iyer, K. A., Bird, R. E., Ivanov, J., Banerjee, S., Georges, G., & Zhou, Q. A. (2024). Review of carbon nanotube research and development: materials and emerging applications. *ACS Applied Nano Materials*, 7(16), 18695–18713. <https://doi.org/10.1021/acsanm.4c02721>
- Ibrahim, M. H., Hamzah, N., Mohd Yusop, M. Z., Septiani, N. L. W., & Mohd Yasin, M. F. (2023). Control of morphology and crystallinity of CNTs in flame synthesis with one-dimensional reaction zone. *Beilstein Journal of Nanotechnology*, 14, 741–750. <https://doi.org/10.3762/bjnano.14.61>
- Kehl, C., Nooteboom, P. D., Kaandorp, M. L. A., & Seville, E. (2023). Efficiently simulating Lagrangian particles in large-scale ocean flows - data structures and their impact on geophysical applications. *Computers & Geosciences*, 175, 105322. <https://doi.org/10.1016/j.cageo.2023.105322>
- Lavagna, L., Nisticò, R., Musso, S., & Pavese, M. (2021). Functionalization as a way to enhance dispersion of carbon nanotubes in matrices: a review. *Materials Today Chemistry*, 20, 100477. <https://doi.org/10.1016/j.mtchem.2021.100477>
- Li, M., Risa, M., Osawa, T., Sugime, H., & Noda, S. (2020). Facile catalyst deposition using mists for fluidized-bed production of sub-millimeter-long carbon nanotubes. *Carbon*, 167, 256–263. <https://doi.org/10.1016/j.carbon.2020.06.018>

- Liu, W., Liu, D., Zhang, Y., & Li, B. (2022). Numerical investigation of particle size distribution, particle transport and deposition in a modified chemical vapor deposition process. *Powder Technology*, 407, 117616. <https://doi.org/10.1016/j.powtec.2022.117616>
- Liu, Y., Vourliotakis, G., Hardalupas, Y., & Taylor, A. M. K. P. (2017). Experimental and numerical study of chemiluminescence characteristics in premixed counterflow flames of methane based fuel blends. 55th AIAA Aerospace Sciences Meeting (pp. 1-12). AIAA Publisher. <https://doi.org/10.2514/6.2017-0153>
- Martin, J. W., Salamanca, M., & Kraft, M. (2021). Soot inception: carbonaceous nanoparticle formation in flames. *Progress in Energy and Combustion Science*, 88, 100956. <https://doi.org/10.1016/j.pecs.2021.100956>
- Meng, B., Zhang, Z. Y., Ma, L. Y., & Wan, M. (2022). Effect of sintering temperature on microstructure and mechanical properties of Inconel 718 superalloy prepared by micro-FAST. *Materials Science and Engineering A*, 836, 142733. <https://doi.org/10.1016/j.msea.2022.142733>
- Muchuweni, E., Mombeshora, E. T., Martincigh, B. S., & Nyamori, V. O. (2022). Recent applications of carbon nanotubes in organic solar cells. *Frontiers in Chemistry*, 9, 733552. <https://doi.org/10.3389/fchem.2021.733552>
- Murjani, B. O., Kadu, P. S., Bansod, M., Vaidya, S. S., & Yadav, M. D. (2022). Carbon nanotubes in biomedical applications: current status, promises, and challenges. *Carbon Letters*, 32(5), 1207–1226. <https://doi.org/10.1007/s42823-022-00364-4>
- Nakazawa, S., Yokomori, T., & Mizomoto, M. (2005). Flame synthesis of carbon nanotubes in a wall stagnation flow. *Chemical Physics Letters*, 403(1-3), 158–162. <https://doi.org/10.1016/j.cplett.2004.12.091>
- Ndayishimiye, A., Fan, Z., Mena-Garcia, J., Anderson, J. M., & Randall, C. A. (2022). Coalescence in cold sintering: a study on sodium molybdate. *Open Ceramics*, 11, 100293. <https://doi.org/10.1016/j.oceram.2022.100293>
- Norizan, M. N., Moklis, M. H., Ngah Demon, S. Z., Abdul Halim, N., Samsuri, A., Mohamad, I. M., Knight, V. F., & Abdullah, N. (2020). Carbon nanotubes: functionalisation and their application in chemical sensors. *RSC Advances*, 10(71), 43704–43732. <https://doi.org/10.1039/d0ra09438b>
- Moore, S. (2022). Reporting on the current carbon nanotubes market. *AZoNano*. <https://www.azonano.com/article.aspx?ArticleID=6313>
- Picheau, E., Amar, S., Derré, A., Pénicaud, A., & Hof, F. (2022). An introduction to the combustion of carbon materials. *Chemistry - A European Journal*, 28(54), e202200117. <https://doi.org/10.1002/chem.202200117>
- Posch, S., Gößnitzer, C., Lang, M., Novella, R., Steiner, H., & Wimmer, A. (2025). Turbulent combustion modeling for internal combustion engine CFD: a review. *Progress in Energy and Combustion Science*, 106, 101200. <https://doi.org/10.1016/j.pecs.2024.101200>
- Somoju, R. (2022). Effect of sintering temperature on microstructure and electrical properties of Ce_{0.82}Dy_{0.155}Sr_{0.025}O_{2-δ} electrolyte. *Processing and Application of Ceramics*, 16(2), 175–182. <https://doi.org/10.2298/PAC2202175R>
- Salah, L. S., Ouslimani, N., Bousba, D., Huynen, I., Danléé, Y., & Aksas, H. (2021). Carbon nanotubes (CNTs) from synthesis to functionalized (CNTs) using conventional and new chemical approaches. *Journal of Nanomaterials*, 2021(1), 4972770. <https://doi.org/10.1155/2021/4972770>

- Sawyer, W. J., & Hart, A. J. (2025). Aerosol synthesis of high-quality single-wall carbon nanotubes through integrated microplasma generation of catalyst nanoparticles. *Chemical Engineering Journal*, 507, 160690. <https://doi.org/10.1016/j.cej.2025.160690>
- Selamat, M. S. R., Zainal, M. T., Yasin, M. F. M., Hamzah, N., & Che Sidik, N. A. (2023). Modelling of the flame synthesis of single-walled carbon nanotubes in non-premixed flames with aerosol catalyst. *Journal of Advanced Research in Numerical Heat Transfer*, 13(1), 39–51. <https://doi.org/10.37934/arnht.13.1.3951>
- Turns, S. R. (2012). *An introduction to combustion: concepts and applications*. (3rd Ed.). McGraw-Hill.
- Wang, H. (2011). Formation of nascent soot and other condensed-phase materials in flames. *Proceedings of the Combustion Institute* (pp. 41–67). Elsevier. <https://doi.org/10.1016/j.proci.2010.09.009>
- Xu, F., Liu, X., & Tse, S. D. (2006). Synthesis of carbon nanotubes on metal alloy substrates with voltage bias in methane inverse diffusion flames. *Carbon*, 44(3), 570–577. <https://doi.org/10.1016/j.carbon.2005.07.043>
- Zainal, M. T., Mohd Yasin, M. F., Wan Ali, W. F. F., Tamrin, K. F., & Ani, M. H. (2020). Carbon precursor analysis for catalytic growth of carbon nanotube in flame synthesis based on semi-empirical approach. *Carbon Letters*, 30(5), 569–579. <https://doi.org/10.1007/s42823-020-00127-z>
- Zainal, M. T., Mohd Yasin, M. F., Abdul Wahid, M., & Mohd Sies, M. (2021). A flame structure approach for controlling carbon nanotube growth in flame synthesis. *Combustion Science and Technology*, 193(8), 1326–1342. <https://doi.org/10.1080/00102202.2019.1694518>
- Zainal, M. T., Hamzah, N., Abdul Wahid, M., Kamaruzaman, N., Chong, C. T., Ani, M. H., Amzin, S., Das, T., & Mohd Yasin, M. F. (2023). Zero-dimensional model for the prediction of carbon nanotube (CNT) growth region in heterogeneous methane-flame environment. *Carbon Letters*, 33(7), 2199–2210. <https://doi.org/10.1007/s42823-023-00579-z>
- Zhang, C., Tian, B., Chong, C. T., Ding, B., Fan, L., Chang, X., & Hochgreb, S. (2020). Synthesis of single-walled carbon nanotubes in rich hydrogen/air flames. *Materials Chemistry and Physics*, 254, 123479. <https://doi.org/10.1016/j.matchemphys.2020.123479>
- Zhang, C., Tian, B., Chong, C. T., Ding, B., Fan, L., Chang, X., & Hochgreb, S. (2022). Experimental study of thiophene and ferrocene in synthesis of single-walled carbon nanotubes in rich premixed hydrogen/air flames. *Combustion and Flame*, 238, 111939. <https://doi.org/10.1016/j.combustflame.2021.111939>
- Zhang, Z., Zhang, N., & Zhang, Z. (2025). High-performance carbon nanotube electronic devices: progress and challenges. *Micromachines*, 16(5), 554. <https://doi.org/10.3390/mi16050554>



© 2026 by the authors. This article is an open access article distributed under the terms and conditions of the Creative Commons Attribution (CC BY-NC-SA) license (<https://creativecommons.org/licenses/by-nc-sa/4.0/>)