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Citation for published version (APA):
Kalbhor, A., & van Oijen, J. A. (2022). Effects of curvature on soot formation in steady and unsteady counterflow diffusion flames. Combustion and Flame, 241, [112108]. https://doi.org/10.1016/j.combustflame.2022.112108

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DOI:
10.1016/j.combustflame.2022.112108

Document status and date:
Published: 01/07/2022

Document Version:
Publisher’s PDF, also known as Version of Record (includes final page, issue and volume numbers)

Please check the document version of this publication:
• A submitted manuscript is the version of the article upon submission and before peer-review. There can be important differences between the submitted version and the official published version of record. People interested in the research are advised to contact the author for the final version of the publication, or visit the DOI to the publisher’s website.
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Effects of curvature on soot formation in steady and unsteady counterflow diffusion flames

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**A R T I C L E   I N F O**

Article history:
Received 24 July 2021
Revised 1 March 2022
Accepted 8 March 2022

Keywords:
Flame curvature
Discrete sectional method
Soot formation
Non-premixed flames

**A B S T R A C T**

A numerical study has been conducted in order to understand the effects of flame curvature on soot formation in laminar non-premixed flames. For the fundamental understanding, the canonical configuration of a counterflow diffusion flame is employed as it exhibits the essential combustion physics associated with non-premixed flames. Numerical results for the steady counterflow ethylene flames confirmed that the response of soot in curved flames is governed by the intricate coupling between flow convection, soot kinetics, and differential transport of soot impacting the rates of soot formation sub-processes. The sensitivity of the soot formation in curved flames to the strain rate variation is also analyzed to understand their competing effects. Furthermore, the dynamic response of soot formation to the unsteady curvature is investigated by imposing harmonic oscillations to the curvature. Numerical results revealed that for an increase in the frequency of curvature oscillations, the soot formation gets attenuated and amplitudes of the induced oscillations exhibit large phase-shift with respect to imposed fluctuations. The higher characteristic time scales of larger-sized particles proved to control the overall dynamic response of soot under unsteady fluctuations of curvature. To examine the dynamic response of soot formation under a more complex case of flow-flame-soot interaction, the unsteady analysis of flames is extended by subjecting them to the simultaneous strain rate and curvature oscillations. The correlation between imposed frequencies and variability of soot formation response under fluctuating strain rate and curvature is elaborated.

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1. Introduction

Owing to the stringent legislative regulations, mitigating the soot emission from combustion devices is a critical challenge in modern-day combustion research. In particular, many practical combustion systems employ non-premixed flames, which generally exhibit a higher sooting tendency compared to premixed flames [1]. Most of these combustion systems operate under conditions characterized by complex unsteady hydro-thermo-chemical interactions. Therefore, understanding the soot formation process in non-premixed flames under a varied range of conditions encountered in such devices is pivotal, especially, for the development of reliable soot modeling approaches.

In turbulent environments, the soot formation is highly influenced by the unsteady hydrodynamics that introduces spatio-temporally fluctuating stretch and curvatures to the flames. On the other hand, in laminar non-premixed flames, where the flame dynamics is not turbulence-induced, curvature effects are also identified [2]. Since flame curvature primarily affects the local flame surface area, it strongly impacts diffusion transport of different chemical species, and therefore, modifies the overall flame structure. Accordingly, in curved non-premixed flames, the flux of species mass is enhanced towards or away from the fuel-rich or lean zones based on the type of curvature (convex/concave) and diffusivity of the chemical species. Since the soot formation process is particularly sensitive to local mixture composition and differential diffusion effects, the non-uniformities of the thermo-chemical variables introduced by the curvature strongly influence the distribution of soot [2,3].

There have been notable studies [3–6] recognizing the effects of flame curvature on soot formation in non-premixed flames. In the context of turbulent flames, previous numerical works [3,4] established that the curvature induces a relative movement of the flame and the soot formation region, which impacts the convection of the soot particles. It was noticed by Lignell et al. [3] that negative flame curvature (for which the center of curvature lies in the fuel side) tends to shift the flame towards the fuel side, which leads
to increased temperatures in the fuel-rich soot growth regions and promotes the soot formation. Positively curved flames, on the other hand, show the opposite behavior leading to a diminished soot concentration. Franzelli et al. [5] investigated the correlation between soot formation rates and the response of soot precursors to curvature in laminar unsteady diffusion flames. Their numerical study demonstrated that a negatively curved flame front manifests an increased convective motion of gas and soot towards the fuel-rich zone. This enhances the transport of heat from the flame towards the preheat region and leads to increased concentrations of PAHs, and consequently, the overall soot production, while the opposite holds for positive curvature. However, specific soot sub-processes were not analyzed concerning the curvature effects. Recently, Hoerle [6] compared the effect of flame curvature on the soot formation in steady laminar counterflow diffusion flames to characterize its influence on flamelet-based models. Although the physical mechanism for the observed steady-state soot response was not explicitly addressed, the enhancement of soot concentration by negative curvature was evident in oxyfuel flames.

In summary, the previous studies show a consensus about the enhancement of soot formation under negative curvature for laminar and turbulent non-premixed flames. However, although various mechanisms have been postulated, the origin of soot response to flame curvature is not well understood. In this context, a comprehensive examination of soot sub-processes with state-of-the-art detailed kinetics and a sectional method-based soot model is useful to encapsulate the mechanism of soot formation and evolution of their size distributions in curved non-premixed flames. Moreover, the impact of curvature unsteadiness on soot production has not been sufficiently addressed. In particular, it would be interesting to characterize the dynamic response of sooting flames subjected to unsteady curvature fluctuations, and identify the underlying physical phenomena which control it.

Nevertheless, from a numerical perspective, analyzing unsteady curvature effects on soot formation directly in multi-dimensional flames is very challenging owing to the co-existence of different physical processes impacting chemistry and local flow properties. Therefore, alternatively, the academic configuration of a counterflow diffusion flame is attractive to acquire fundamental insight into the physics and kinetics occurring in non-premixed flame fronts under the influence of curvature [6]. Furthermore, by employing a counterflow configuration, the dynamic response of soot to the unsteady curvature could be exemplified by systematically exposing laminar flames to harmonically varying curvature over a range of frequencies and amplitudes. Although the fundamental studies on canonical configurations do not reproduce the complexities encountered in fully turbulent simulations, they do provide crucial information to recognize the effects of unsteady curvature fluctuations on the soot formation process and guide the modeling efforts. For instance, the importance of including curvature effects in flamelet-based modeling has been recognized in the literature [2,6,7]. In this context, the analysis of curvature unsteadiness on soot formation would provide important insights into the development of flamelet-based models for application to multi-dimensional non-premixed sooting flame simulations.

Furthermore, in practical burners, flames are subjected to both varying curvatures and strain rates. As a result, the soot formation process is strongly influenced by the changes in the strain rate. The effects of strain rate on the soot and PAH formation in non-premixed flames have been addressed in several experimental [8–10] and numerical works [11–14]. In general, previous studies showed that an increase in strain rate leads to a reduction in soot formation. Such an effect is attributed mainly to the lowered concentration of soot precursors leading to reduced soot inception rates, and reduced residence time for soot surface reactions and particle-particle interactions. Moreover, strain rate fluctuations significantly affect the local flow time scales [12,15]. Therefore, to envisage a more complex scenario of flow-flame-soot interaction, it would be interesting to investigate the combined effects of strain and curvature on soot characteristics and the sensitivity of soot formation in curved flames exposed to varying strain rates.

In view of the above considerations, the main objectives of the present study are:

1. to conduct a detailed analysis of different soot sub-processes and provide enhanced insight into the mechanism governing soot response in a laminar counterflow diffusion flame under the influence of flame curvature.
2. to examine the sensitivity of soot formation in curved flames to strain rate and evaluate the existence of synergistic effects.
3. to systematically characterize the unsteady dynamics of soot formation in a counterflow diffusion flame subjected to curvature oscillations and identify the underlying mechanism.
4. to investigate the competing effects of simultaneously imposed curvature and strain rate fluctuations on the dynamic response of soot.

To the authors’ knowledge, this is the first study that investigates the dynamic response of soot in counterflow diffusion flames under unsteady curvatures. Hence, the findings of the current study provide a fundamental understanding of the curvature effects on soot formation which would be relevant to soot modeling in multi-dimensional flame simulations.

The paper is organized as follows: In Section 2 the numerical methodology and soot model are described briefly. In Section 3 the analysis of curvature effects under steady conditions is conducted for the soot formation (SF) type flame. Section 4 presents the analysis concerning the dynamic response of soot formation to unsteady strain rate and curvature oscillations. Concluding remarks are summarized in Section 5.

2. Numerical methodology

2.1. Governing equations

The numerical simulations are carried out using the one-dimensional laminar flamelet solver CHEM1D [16,17]. A system of governing equations for conservation of total mass, species mass, and enthalpy can be written in a Cartesian system along the coordinate (x) normal to flame surface as:

\[
\frac{\partial \rho}{\partial t} + \frac{1}{\zeta} \frac{\partial (\zeta \rho u)}{\partial x} = - \rho K
\]  
(1)

\[
\frac{\partial (\rho Y_k)}{\partial t} + \frac{1}{\zeta} \frac{\partial (\zeta \rho u Y_k)}{\partial x} = - \frac{1}{\zeta} \frac{\partial (\zeta \rho V_k V_s)}{\partial x} - \rho K Y_k + \omega_k, \ \forall k \in [1, N_s - 1]
\]  
(2)

\[
\frac{\partial (\rho h)}{\partial t} + \frac{1}{\zeta} \frac{\partial (\zeta \rho u h)}{\partial x} = - \frac{1}{\zeta} \frac{\partial (\lambda \frac{\partial h}{\partial x})}{\partial x} - \frac{N_s}{\zeta} \sum_{k=1}^{N_s} \rho h_k Y_k V_s - \rho K h
\]  
(3)

where \( Y_k, \omega_k, h_k, \) and \( V_s \) are, respectively, mass fraction, chemical source term, enthalpy, and the diffusion velocity of species \( k \). Mixture density, total enthalpy, and \( x \)-directional flow velocity are given by \( \rho, h \) and \( u \), respectively. Mixture conductivity and specific heat capacity are, respectively, denoted by \( \lambda \) and \( c_p \).

The variable \( \zeta \) in the equations reported above indicates the measure for the flame surface through which the mass transport takes place. The surface area function \( \zeta \) is related to the curvature of flame \( \kappa \) as:

\[
\kappa = - \frac{1}{\zeta} \frac{\partial \zeta}{\partial x}
\]  
(4)
When $\kappa$ is independent of $x$, this yields

$$\zeta = \exp(-\kappa x).$$  \hspace{1cm} (5)

To remove the translational degree of freedom, the stagnation plane (where $u=0$) is specified at $x=0$. In the present work, negative curvature indicates that the center of curvature lies on the fuel side ($x \to -\infty$ here), and the converse is true for positive curvature. It implies, $\zeta$ increases moving from fuel to oxidizer side along the flame coordinate $x$ for negatively curved flames, while the opposite occurs for positively curved flames. A schematic diagram of the sign convention for the flame curvature followed in the present study is presented in Fig. 1.

In the quasi-one dimensional formulation of the counterflow configuration, to account for the flow component (with $v$ velocity) in the direction tangential ($\gamma$) to the flame surface, the strain field $K(x, t)$ is defined as:

$$K = \frac{\partial v}{\gamma}. \hspace{1cm} (6)$$

The determination of strain field solution requires an additional $\gamma$-momentum equation. Following the approach of Dixon-Lewis [18], the $\gamma$-momentum equation can be used to derive an equation for $K$ as:

$$\frac{\partial (\rho K)}{\partial t} + \frac{1}{\zeta} \frac{\partial (\zeta \rho u K)}{\partial x} = \frac{1}{\zeta} \frac{\partial}{\partial x} \left( \mu \frac{\partial K}{\partial x} \right) - \rho K^2 + J \hspace{1cm} (7)$$

where $\mu$ is the dynamic viscosity, and $J$ is the pressure curvature in the $\gamma$-direction, i.e.

$$J = \frac{-1}{\gamma} \frac{\partial p}{\partial \gamma}. \hspace{1cm} (8)$$

To obtain $J$, a potential flow approximation is considered in the farfield. Accordingly for the oxidizer stream ($x \to +\infty$) a velocity gradient can be prescribed, such that:

$$\frac{\partial u}{\partial x} = -\frac{\partial v}{\partial \gamma} = -K = -a; \hspace{1cm} (x \to +\infty) \hspace{1cm} (9a)$$

$$\frac{\partial K}{\partial x} = 0; \hspace{1cm} (x \to -\infty) \hspace{1cm} (9b)$$

where $a$ denotes the applied strain rate at the oxidizer side. From Eq. 7 and Eq. 9 the expression for $J$ becomes:

$$J = \rho_o a^2 \hspace{1cm} (10)$$

with $\rho_o$ the density of oxidizer.

For the closure of the conservation equations, the low Mach-number approximation for the equation of state is used. Heating due to viscous dissipation, radiative heat transfer, Soret, and Dufour effects are neglected. A mixture-averaged approximation of Curtis and Hirschfelder [19] is used for modeling the diffusion transport of gas-phase species. It is worth highlighting that the counterflow diffusion flames are computed in physical space ($x$), instead of mixture fraction ($Z$) space. The details regarding the derivation and assumptions of the above governing equations can also be found in Refs. [20–22].

2.2. Soot model

A discrete sectional method-based soot model described in the work of Hoerle and Pereira [23] is utilized to predict the soot formation characteristics. In the soot model, a range of soot particle sizes is discretized into a finite number of sections and representative volumes are assigned to individual sections. The transport equations are solved for sectional soot mass fraction $Y_{s,i}$ by accounting for flow convection, thermophoresis, and soot source terms in the form similar to flamelet equations as:

$$\frac{\partial (\rho Y_{s,i})}{\partial t} + \frac{1}{\zeta} \frac{\partial}{\partial x} \left( \zeta \rho u Y_{s,i} \right) = \frac{1}{\zeta} \frac{\partial}{\partial x} \left( \mu \frac{\partial Y_{s,i}}{\partial x} \right) - \rho V_T Y_{s,i} + \rho D_{s,i} \frac{\partial Y_{s,i}}{\partial x} - \rho K Y_{s,i} + \dot{\omega}_{s,i}, \hspace{1cm} \forall \ i \in [1, N_{sec}] \hspace{1cm} (11)$$

where $V_T$ is the thermophoretic velocity of the soot particles specified in [24], $D_{s,i}$ is the diffusion coefficient, and $\dot{\omega}_{s,i}$ is the rate of the total soot mass growth in section $i$. The total sectional soot mass growth rate is computed by accounting for the contributions of soot nucleation, PAH condensation, surface growth, oxidation, and coagulation sub-processes. The soot nucleation is modeled based on pyrene (AA) dimerization. The PAH condensation is assumed to occur through Brownian collisions and coalescence of PAH (AA here) on the soot particle surface. The surface growth and oxidation of soot particles are modeled through the standard hydrogen abstraction acetylene addition (HACA) mechanism [25]. Soot oxidation by OH and O$_2$ is considered. The coagulation model of Kumar and Ramkrishna [26] is used to describe the soot particle dynamics. The present soot model relies on the mono-variate (volume-based) approximation of the sectional method with spherical soot particles in each representative section. For simplicity, the heat loss through soot radiation is not included in the current model.

It is worth highlighting that the curvature is an external effect induced by the flow which affects the flame structure. Therefore, the explicit development/adjustment of soot models in order to capture curvature effects is not necessary, especially when examining the qualitative aspects. Moreover, the sectional method-based soot models have been applied previously in laminar coflow flames wherein curvature effects are introduced either by the flow vortex [5] or due to geometry of the burner configuration [27–29]. The sectional soot models have also been validated in investigating transient dynamics of soot [12,13,15]. Therefore, for the present study, it is recognized as the appropriate soot model choice.

2.3. Target flame

To demonstrate the effects of curvature on the sooting characteristics, the counterflow diffusion flame studied by Wang et al. [30] is chosen as the flat baseline case. In this flame, the fuel is pure ethylene ($C_2H_4$), and the oxidizer composition consist of 25%/75% of O$_2$/N$_2$ (on volume basis). Both fuel and oxidizer streams are maintained at atmospheric pressure and 300K temperature. This flame is an example of a SF (soot-formation) type [31], in which the flame front (characterized by the stoichiometric mixture fraction $Z_{st}$) resides on the oxidizer side of the stagnation plane. Therefore, soot particles are mainly convected away from the flame reaction layer towards the stagnation plane. The retained soot model has been validated for this flame in our previous
work [32]. The detailed scheme KM2 of Wang et al. [33], involving 202 species and 1351 reactions, is used in the present work for the gas-phase chemistry calculations.

3. Simulations of steady flames

Prior to investigating unsteady curvature effects, which is central to the present work, it is important to identify the soot response to curvature in steady-state conditions to facilitate a clear discussion.

3.1. Effects of curvature on soot formation in a steady flame: Baseline case

First, we briefly discuss the effects of flame curvature on the soot formation characteristics for the steady baseline flame. To isolate the effects of curvature, the strain rate is kept constant ($\dot{\alpha}=120 s^{-1}$), and a curvature of $\kappa=\pm 5cm^{-1}$ is applied. The value of selected $\kappa$ for the baseline flame is relatively moderate and relates to the diffusion flame thickness $\delta_{f,Z}$ such that:

$$\kappa | \delta_{f,Z} = \mathcal{O}(1)$$

in which, the diffusion flame thickness $\delta_{f,Z}$ is approximated as [34]:

$$\delta_{f,Z} \approx \frac{2Z_{st}}{\nabla Z_{st}}.$$  \hspace{1cm} (13)

The mixture fraction definition here follows from the work of Bilger [35]. For the baseline counterflow flame considered, Eq. 13 yields, $1/\delta_{f,Z} \approx 4.6cm^{-1}$.

Prior to discussing the soot characteristics, it is important to illustrate key features of the flame structure under the influence of curvature. In particular, since the flame curvature causes a relative change in the local area of the flame surface, it alters the total mass flux ($m = \rho u$) of the reactants. Fig. 2a demonstrates that a negative curvature enhances the mass flux of fuel towards the stagnation plane as a consequence of the reduction in the local flame surface area, while positive curvature causes the opposite effect, and increases the mass flux of oxidizer towards the stagnation plane. The change in mass flux strongly impacts the local mixing characteristic of the flame through the differential transport between the gas-phase species and mixture fraction [2]. The movement of mixture fraction iso-surfaces owing to curvature effects is evident from Fig. 2b. A negative curvature tends to shift the reaction zone towards the fuel side, as a result, the mixture fraction at the flame stagnation position is found to be decreased compared to its flat flame counterpart. Positive curvature, on the other hand, shows the opposite effect. As will be shown later, for the target flame under study, the soot formation zone (characterized by the rate profiles of different soot sub-processes) encompasses the composition space from about $Z \sim 0.15$ to 0.3. From Fig. 2b, it can be noticed that the soot formation zone shift towards the fuel side of the stagnation plane for negative curvature. On the contrary, the soot formation zone is found to shift towards the oxidizer side for positive curvature. Moreover, flame curvature also impacts the distribution of temperature in physical space (see Fig. 2c). For negative curvature, the reaction zone tends to shift towards the fuel side, enhancing the transport of heat from the flame towards the fuel diffusive layer [5]. On the contrary, for the applied value of positive curvature, the temperature profile shifts towards the oxidizer side. Such response of temperature to curvature is mainly attributed to preferential diffusion effects [36].

Given the curvature-induced displacement of mixture fraction, the analysis of the soot formation process in physical space is relatively intricate. Therefore to elucidate the curvature effects on soot, in Fig. 3, profiles of global soot quantities are presented in $Z$-space. From the numerical results, it is evident that negative curvature increases the peak value of the soot volume fraction ($f_v$), whereas the same is found to be decreased when positive curvature is applied. Furthermore, it can be observed that negative curvature tends to shift the peak of $f_v$ towards the lower mixture fraction while making its profile narrower in $Z$-space compared to the
flat flame. On the contrary, positive curvature leads to a wider $f_v$ profile with a peak shifting towards higher $Z$. The observed qualitative response of $f_v$ under curvature is in agreement with the previous works \cite{5,23}. The density change also contributes to the increasing $f_v$ towards the stagnation plane. To isolate the influence of density change from the $f_v$, the profiles of soot mass fraction $Y_s$ are also compared in Fig. 3b. As can be seen, the qualitative trends of $Y_s$ with curvature follow the trend in $f_v$. Moreover, Fig. 3c shows that, compared to a flat flame, negative curvature causes a stronger decay in the soot number density ($N$) as soot particles grow and convect towards the stagnation plane, while the opposite tendency occurs for positive curvature. The lowered number density accompanied by a larger average soot particle diameter $d_{avg}$ (in Fig. 3d) mainly reflects the enhanced growth rate of soot particles due to negative curvature. In contrast, the soot particle size tends to decrease under positive curvature, implying a relatively high number density of smaller-sized particles. It is worth highlighting that a small second peak (note the log scale) in number density is found at higher $Z$. This is caused by the small amount of A4 predicted by the employed KM2 mechanism, which leads to nucleation. However, growth rates of subsequent soot sub-processes are negligible there, and therefore, the soot mass fraction remains small.

In general, the process of soot formation is strongly influenced by the complex flow-flame interactions, which affect the residence time of soot particles, thermophoresis, and soot chemistry. Therefore, to understand the physical mechanism associated with observed soot response in curved flames, a detailed analysis of the aforementioned phenomena is essential. As mentioned earlier, due to the change in flame surface area, the flame curvature significantly alters the flow characteristics. Since the soot formation is a relatively slow process, it is interesting to investigate the impact of curvature on its residence time. To that end, the profiles of soot particle velocity ($\text{flow velocity } u$ combined with the thermophoretic component $V_f$) are plotted against mixture fraction in Fig. 4. It can be seen that negative curvature substantially reduces the soot particle velocity compared to that in a flat flame. The lowered velocity implies an enhanced residence time of soot particles as they grow and convect towards the stagnation plane. Therefore, the observed increment in $f_v$ by negative curvature can be partially attributed to the higher soot residence time, which contributes to the increased soot mass growth. On the other hand, for positive curvature, the soot particle velocity is higher compared to a flat flame, which tends to lower the residence time of soot, and consequently lowering its overall growth.

It is well-known that thermophoresis strongly affects the transport of soot particles. Therefore, it is useful to examine the impact of curvature on its contribution to the overall soot response. To address this, the profiles of soot volume fraction in curved flames without thermometric diffusion ($V_f = 0$) are compared in Fig. 5. In SF type flames, thermophoresis shifts $f_v$ profiles towards higher $Z$ values while reducing their peaks. Naturally, it can be observed that the peak values of $f_v$ are significantly altered by neglecting thermophoresis. However, it does not impact the qualitative trends in $f_v$ profiles with curvature. Hence the role of thermophoretic diffusion on soot response in curved flames appears to be secondary.

Next, to assess the influence of curvature on soot chemistry, the rate profiles of different sub-processes are presented in Fig. 6. As can be observed, rates of PAH condensation, surface growth, and oxidation are enhanced by negative curvature and diminished by positive curvature. On the other hand, the impact of curvature on soot nucleation is somewhat complex. The nucleation rates show lowered peak values for both negative and positive curva-
tures compared to the flat flame. Since, in the present model, the soot is assumed to nucleate through the dimerization of A4 molecules, the profiles of A4 mass fraction are compared in Fig. 7a. The decreased rates of soot nucleation in curved flames are recognized to be a direct consequence of the A4 concentrations. Note that the profiles of A4 presented in Fig. 7a include the consumption through soot sub-processes. Therefore, pure gas-phase results (without soot model) are also analyzed (see Fig. 7b) to verify the trends in A4 profiles under curvature effects. The gas-phase results for curved flames (irrespective of the sign of the curvature) also do not show enhanced A4 mass fraction peaks compared to the flat flame. Nevertheless, in the soot formation zone ($Z \sim 0.15$ to 0.3), the A4 mass fraction predicted by the gas-phase simulation for the negatively curved is higher compared to the positively curved flame. On the contrary, in sooting flame simulations (see Fig. 7a) A4 mass fraction are found to be only slightly different. This behavior could be related to increased consumption of A4 through enhanced PAH-condensation rates under negative curvature. Therefore, from the analysis of A4 profiles, the direct causal relationship between PAH and the observed soot response under curvature is not observed. On the other hand, since the rates of surface growth (see Fig. 6) are approximately an order of magnitude greater than nucleation and condensation rates, it is clear that the surface reactions predominantly account for the overall soot mass growth rate.

Fig. 4. Distribution of the soot particle velocity ($u + V_T$) for curved and flat flames.

Fig. 5. Distributions of $f_v$ for curved and flat flames when thermophoretic diffusion is neglected ($V_T = 0$). For comparison, $f_v$ profiles for $V_T \neq 0$ are shown with lighter curves.

Fig. 6. Rate profiles of soot nucleation (a), PAH condensation (b), HACA surface growth (c), and soot oxidation (d) for curved and flat flames.
Fig. 7. Profiles of A4 mass fraction with (a), and without (b) soot kinetics, and C2H2 mass fraction (c) for curved and flat flames.

Fig. 8. Distributions of soot number density $N$ in Z-space for different particle sizes $d_p$. Vertical lines indicate: stagnation plane (cyan), soot zone (white) and stoichiometric Z (yellow). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

in SF type flames. Therefore, the increased soot volume fraction for negative curvature can be attributed to the enhanced rate of soot surface growth.

To explicate the enhanced rates of surface growth in negative curvature, it is important to highlight the crucial role of differential diffusion of soot relative to the gas-phase (mixture fraction).
The key role of differential transport of soot particles in curved non-premixed flames has also been recognized in literature [3,4]. Due to their high Schmidt numbers (owing to lower diffusivities), the transport of soot particles in mixture fraction space is strongly affected by the curvature effects. Analogous to gas-phase species, the contribution of curvature in the convection velocity ($u^*$) of soot mass fraction in $Z$-space is primarily related to [2,37]:

$$u^* = \left(1 - \frac{Le_2}{Le_1}\right) \kappa \sqrt{\frac{\chi D_2}{2}}$$

(14)

where $Le_2, Le_1$ are the Lewis numbers for mixture fraction and soot particles, respectively, while $\chi$ and $D_2$ denote scalar dissipation rate and thermal diffusivity, respectively. A detailed description of the flamelet equations in $Z$-space can be found elsewhere [2,37]. Negative curvature, therefore, induces the convection of soot particles ($Le_3 \gg Le_2$) towards lower mixture fraction. As a result, soot particles are differentially transported (relative to mixture fraction) towards lower $Z$ values.

A more visual illustration of the differential transport of soot particles is presented through the sectional distribution of particle number density in Fig. 8. It depicts the number density of the different particle sizes as a function of $Z$. It is evident from Fig. 8a that under negative curvature, a large number of soot particles are found within the soot formation zone (region of higher rate profiles of sub-processes). As a result, the particles grow and the number density of larger-sized soot particles within the soot formation zone is increased. In contrast, positive curvature tends to transport soot particles towards higher $Z$, causing them to move away from the soot formation zone, reducing their growth rate. Under the HACA mechanism, employed in the present work, the rate of surface growth primarily depends on the concentrations of C$_2$H$_2$ and active surface radical sites. The concentration of active surface radicals is directly related to the size and number density of soot particles. From the mass fraction profiles of C$_2$H$_2$ (Fig. 7c), it is apparent that the peak C$_2$H$_2$ concentration is hardly affected by flame curvature. Therefore, the increment in the surface growth rate by negative curvature is essentially a consequence of the increased concentration of surface sites facilitated by the curvature-induced transport of soot particles in the sooting zone.

Moreover, the soot particles grown through surface reaction and condensation, coagulate as they approach the stagnation plane. Naturally, the increased concentration of larger-sized soot particles by negative curvature also impacts the process of soot coagulation. To illustrate this, the distributions of coagulation rates of different sized particles in mixture fraction space under curvature are compared in Fig. 9. The enhanced coagulation rates towards larger-sized particles can be noticed for negative curvature. The increased coagulation rates of larger-sized particles subsequently lead to larger particle diameters, which further promote surface reactions through the increased surface area density of soot particles. Since negative curvature tends to increase the soot surface area density, the soot oxidation rates are also enhanced (see Fig. 6d). However, the contribution of the soot oxidation process in the overall soot production is insignificant compared to surface growth. On the contrary, the lowered flux of soot particles in positively curved flames adversely impacts the coagulation rates, contributing to lower soot volume fraction.

In summary, the overall response of soot to curvature effects appears to be governed by two key physical mechanisms that complement each other. For a negatively curved flame, lowered particle velocities tend to enhance the residence time of soot. In addition, the differential transport of soot particles towards the soot formation zone leads to increased surface growth rates. In positively curved flames, the exact opposite behavior is noticed, which results in lowered soot volume fractions.

The use of the sectional method-based model enables the analysis of soot particle size distribution. In this context, the influence of flame curvature on the particle size distribution function (PSDF) at the peak $f_v$ position is shown in Fig. 10. PSDF profiles show a bimodal behavior in the flat as well as curved flames. For a negative curvature, the response of PSDF in log-normal mode is driven by the increased rate of surface growth and enhanced rate of coagulation between larger-sized particles, which leads to the shifting of the peak in the log-normal mode of PSDF towards larger particle diameters. The opposite behavior can be noticed in the positively curved flames as a consequence of diminished rates of surface growth and coagulation. On the other hand, negative curvature shows a higher number of incipient particles respective to the flat flame in a power-law mode. This is essentially a reflection of a somewhat higher local rates of nucleation compared to its flat and positive curvature counterparts (see Fig. 6a). Note that the peak $f_v$ positions for curved flames are shifted in $Z$-space. While the maximum rate of nucleation were observed to reduce for negative curvature (Fig. 6a), the nucleation rates at the location of peak $f_v$ shows a slight increase. Moreover, compared to a flat flame, nega-
itive curvature shifts the trough in the PSDF towards the larger particle diameters \( (d_p) \) while making it shallower with an increment in number density. Note that this discussion of the PSDF only represents qualitative tendencies as soot agglomeration and fragmentation processes are not explicitly considered in the present model.

Next, to examine the sensitivity of soot formation processes to flame curvature, additional simulations are carried out for curvature values ranging from \(-10 \text{ cm}^{-1}\) to \(10 \text{ cm}^{-1}\). The variations of normalized peak soot volume fraction and peak soot formation rates are depicted against curvature in Fig. 11. The peak quantities are normalized by their corresponding values for a flat flame. Fig. 11 reveals that the peak soot volume fraction increases monotonically from positive to negative curvatures. In other words, soot formation tends to enhance for a negative curvature, and mitigate for a positive curvature. However, the relative increment in peak soot volume fraction for negative curvatures is almost similar to its decrement at positive curvatures. Furthermore, the rates of soot surface growth and oxidation show a monotonic increment as curvature decreases from positive to negative values. On the other hand, peak soot nucleation rates and condensation rates do not show a monotonic response to curvature variation. Such a non-monotonic behavior of soot nucleation rates essentially stems from the response of \( A_4 \) to the curvature. For instance, it is noticed that the peak value of \( A_4 \) initially increases for negatively curved flames, attains a maximum, and then again tends to decrease at higher values of negative curvatures. In contrast, for positive curvatures, the peak concentration of \( A_4 \) shows a monotonic decrement with curvature leading to a reduction in soot nucleation rates.

### 3.2. Effects of curvature on soot formation in steady flames: Variation of strain rate

It is known from previous works \([11, 14, 38]\) that at higher strain rates, soot formation is reduced primarily as a consequence of the decreased concentration of gas-phase precursors, and lowered residence time for the growth of soot particles through the coagulation process. To present a more complex picture of flow-flame-soot interactions, the effects of curvature on soot formation characteristics are investigated for different strain rates. In Fig. 12, the peak \( f_s \) are plotted as a function of applied strain rate \( a \) for several \( \kappa \) values. An exponential decrease of soot volume fraction with increasing strain rate is apparent at every \( \kappa \) value. Moreover, the dependence of \( f_s^\text{max} \) reduction on the strain rate remains almost similar for the covered range of \( \kappa \).

To elucidate the sensitivity of soot formation to combined strain rate and curvature variation for steady flames, the normalized (with baseline flat flame) peak soot volume fractions \( (f_s^\text{max}) \) are compared in Fig. 13 over a range of strain rates. A decrease in \( f_s^\text{max} \) with strain rate is evident in Fig. 13a. Furthermore, the enhancement in \( f_s^\text{max} \) by negative curvature, and reduction by positive curvature are clearly noticed Fig. 13a at different strain rates as well.

Since variations in strain rate and curvature can either enhance or diminish the overall soot formation, it would be interesting to evaluate if there exist synergistic effects in terms of soot formation when the flame is curved and strained simultaneously. To quantify such synergistic effects of combined strain and curvature variation on soot formation, we define a synergy parameter \( (\Phi) \) as:

\[
\Phi = \frac{\hat{f}(\kappa, a) - \hat{f}_v(\kappa) \cdot \hat{f}_b(a)}{\hat{f}_v(\kappa) \cdot \hat{f}_b(a)}
\]

with

\[
\hat{f}(\kappa, a) = \frac{f_s^\text{max}(\kappa, a)}{f_s^\text{max}(0, a_0)}
\]

\[
\hat{f}_v(\kappa) = \frac{f_s^\text{max}(\kappa, a_0)}{f_s^\text{max}(0, a_0)}
\]

\[
\hat{f}_b(a) = \frac{f_s^\text{max}(0, a)}{f_s^\text{max}(0, a_0)}
\]

where \( \hat{f}(\kappa, a) \) and \( \hat{f}_v(\kappa) \) respectively, denote the fractional change in \( f_s^\text{max} \) for the variation in curvature at a reference strain rate \( a_0 = 120 \text{s}^{-1} \), and for the variation in strain rate of the flat flame \( (\kappa = 0) \). The first term, \( \hat{f}(\kappa, a) \) in the \( \Phi \) expression refers to the actual change observed in the \( f_s^\text{max} \) compared to the baseline case \( (a = a_0, \kappa = 0) \). On the other hand, the second term signifies the expected change in \( f_s^\text{max} \) as a consequence of variation in curvature and strain rate by considering their individual effects through multiplication.

The values of the synergy parameter \( (\Phi) \) observed under negative and positive curvatures are plotted against the strain rate in Fig. 13b. The analysis of the synergistic response suggests that the sensitivity of an increment in \( f_s^\text{max} \) by negative curvature is enhanced on lowering the strain rate. Positive curvature, on the other hand, hardly shows any significant sensitivity of \( f_s^\text{max} \) reduction to strain rate. Moreover, the synergistic effects are found to be substantially lowered in negatively curved flames for an increment in strain rate. Such an effect can be partially attributed to the decrease in flame thickness \( \delta_f \) at higher strain rates, causing a reduction in dimensionless curvature \( \kappa \delta_f \), which essentially lowers the tendency of the flame structure to be affected through curvature effects [6].
4. Simulations of unsteady flames

In this section, we analyze the dynamic response of soot formation in the target flame to unsteady curvature. The curvature unsteadiness is introduced with a sinusoidal function of the form:

\[ \kappa = \Delta \kappa \cdot \sin(2\pi f_s t) \]  

where \( \Delta \kappa \) is the amplitude, and \( f_s \) the frequency of the curvature oscillations. In addition, to understand the effects of simultaneous strain-curvature oscillations on the soot formation, sinusoidal type fluctuations are imposed on the applied strain rate. The strain rate fluctuations are assumed to have a form:

\[ \dot{a} = a_0 + \Delta a \cdot \sin(2\pi f_s t) \]  

where \( \Delta a \) is the amplitude, and \( f_s \) the frequency of the imposed strain rate oscillations, and \( a_0 = 120s^{-1} \) is the strain rate of the steady-state flame solution. Initially, the flame is planar and at a steady-state, upon which the subsequent oscillations are imposed. At \( t = 0 \), the curvature and strain rate begin to fluctuate according to Eq. 17 and Eq. 18, respectively. It was noticed in [12] that the response of soot and PAH to the imposed strain rate fluctuation is quasi-steady for low frequencies. At higher frequencies, on the other hand, the time scales of soot formation become longer than the time scales of flow field fluctuations, and thus the imposed oscillations could not modify the response of the soot formation process. In the present analysis, the frequencies chosen for strain and curvature fluctuations are in the range of 1-1000Hz. Furthermore, \( \Delta \kappa \) and \( \Delta a \) are selected such that for the limits of unsteady curvature and strain rate variation, steady-state results are also available. To quantify the unsteady effects of separate, and simultaneous curvature and strain rate fluctuations, a wide range of conditions are simulated.

4.1. Dynamic response of soot to fluctuating curvature at constant strain rate

4.1.1. Variation in the frequency of imposed oscillations

Because of its relatively large characteristic time scale, the soot response is very sensitive to the flame unsteadiness [12]. Therefore, unsteadiness imposed through curvature oscillations is expected to strongly affect the dynamic behavior of soot. To elucidate this, the dynamic response of peak soot volume fraction (normalized with the steady-state value) at different values of \( f_s \) is presented in Fig. 14. For reference, the imposed profiles of curvature oscillation are also shown. The time is non-dimensionalized with the period \( (\tau = f_s t) \) of the imposed curvature oscillations. Note that the transient simulations were conducted until the limit cycle is reached. However, for the readability of figures, the results for the \( f_v^{\text{max}} \) response are conveniently presented for the first few periods.

As can be noticed from Fig. 14a, the dynamic response of soot is strongly influenced by the frequency of the imposed curvature oscillations. The amplitude of the induced oscillations in peak soot volume fraction rapidly decreases with an increase in the imposed frequency. This essentially indicates that at higher frequencies, the dynamic response of soot tends to get attenuated. The amplitude variation also exhibits strong asymmetric behavior about the steady-state value. Furthermore, the phase-shift observed in the \( f_v^{\text{max}} \) response is negligible for \( f_s = 1\text{Hz} \), indicating the quasi-steady type response. On the contrary, with an increase in \( f_s \), the phase-lag between imposed oscillations and soot volume fraction response becomes substantial.

To enable a clear comparison with the steady-state response, the transient variation of normalized \( f_v^{\text{max}} \) within an induced oscillation cycle is represented against the instantaneous curvature in Fig. 15. The dotted gray curve in the plot refers to the \( f_v^{\text{max}} \) values obtained in steady-state conditions. The large departure from the steady-state conditions, indicating decoupling between imposed oscillations and the soot response, is more evident at higher \( f_s \). In other words, at high frequency, the curvature fluctuations have a very small effect on the induced amplitude of \( f_v^{\text{max}} \) response. As a consequence, \( f_s = 1000\text{Hz} \) shows no dynamic effects on the soot formation. In contrast, the deviation from the steady-state trajectory becomes insignificant as the imposed frequency is lowered to 1Hz, indicating a quasi-steady response.

The soot response to sinusoidal curvature fluctuations can be further illustrated by comparing soot profiles at different temporal positions. Figure 16 depicts the instantaneous profiles of soot volume fraction at four phases \( (\phi) \) of the limit cycle for different \( f_s \), along with the quasi-steady flame solution (gray symbols). These phase positions correspond to: zero fluctuation and increasing curvature \( (\phi = 0) \), maximum curvature \( (\phi = \pi/2) \), zero fluctuation and decreasing curvature \( (\phi = \pi) \), and minimum curvature \( (\phi = 3\pi/2) \) (refer to Fig. 14b). It is evident from Fig. 16 that the curvature unsteadiness strongly modifies the shape of the soot profile during the oscillations. From soot volume fraction profiles it can be observed that, within a period of oscillation, the peak of \( f_v \) is periodically formed in the soot formation zone \( (2 \sim 0.15 \text{ to } 0.3) \) and transported towards the stagnation plane, where it is convected away by the stretch. It is important to reflect on the fact that, the imposed oscillations in curvature tend to induce fluctuations in the convective and diffusive transport of soot. Therefore, unsteady curvature is expected to have a strong impact on the overall soot formation process. As the frequency of imposed oscillations is increased, the rate at which the soot formation process responds is...
somewhat reduced as the time scales associated with the faster oscillations become smaller than the characteristic time scales of soot, leading to a phase-lag. Such phase-lagged response of peak \( f_v \) at higher \( f_o \) can be noticed from its relative positions in \( Z \)-space during the evolution.

The mechanism governing the dynamic behavior of soot under unsteady curvature can be better explained by comparing the response of the sectional soot mass growth rates. It is also important to emphasize that the characteristic time scales for soot chemistry also increase with the size of soot particles [15]. Therefore, the transient variation of normalized peak soot mass growth rates for soot particles with different sizes (\( \omega_{s,t}^{\text{max}} \)) is presented in Fig. 17. It is clear from the results that the phase-lag and damping noticed in the \( f_v^{\text{max}} \) (Fig. 14a) oscillations is mainly correlated to the dynamic response of \( \omega_{s,t}^{\text{max}} \) at higher sections. Since larger-sized soot particles mainly contribute to the overall soot volume fraction, the dynamic response of peak \( f_v^{\text{max}} \) is predominantly affected by the unsteady behavior of larger particles under the imposed curvature oscillations. Rodrigues et al. [15] reported that the characteristics time scales of the collision phenomena such as coagulation and condensation increase with particle size. Owing to their large characteristic time scales, larger-sized soot particles do not get sufficient time to respond to the rapid changes in the unsteady hydodynamics of the diffusion flame, induced by the imposed high-frequency curvature fluctuations. This essentially leads to a time-shift between the imposed curvature oscillations and induced soot oscillations, resulting in the larger phase-lag. When the curvature fluctuations are imposed at lower frequencies, the changes in convective and diffusive transport are more prominently felt by the larger-sized soot particles. The soot formation process in the larger sections then responds to the slowly evolving hydodynamics in a quasi-steady-like manner, leading to smaller phase-lag and smaller damping, as evident in Fig. 17.

To quantitatively illustrate the arguments discussed above, in Fig. 18, the phase-lag between the quasi-steady and induced oscillations of \( \omega_{s,t}^{\text{max}} \) for different sized particles is compared against the phase-lag observed in the global \( f_v^{\text{max}} \) response at different \( f_v \) values. It can be observed that the phase-lag increases with the soot particles’ size, and with the increase in the imposed frequency, the phase-lag for a particular size of particles, enhances. Furthermore, it is evident from the results that the phase-lag observed in the \( f_v^{\text{max}} \) response is approximately equivalent to the phase-lag noticed in the \( \omega_{s,t}^{\text{max}} \) response of larger-sized particles. Therefore, it can be affirmed that the dynamic response of \( f_v^{\text{max}} \) at unsteady curvatures in terms of phase and amplitude maintain a hierarchical relationship with the dynamic response of the sectional soot mass growth rates. Nevertheless, the phase-lag observed in the \( \omega_{s,t}^{\text{max}} \) response...
of \(d_p \approx 86\text{nm}\) under 200Hz is significantly higher than the phase-lag for \(f_{\phi}^{\text{max}}\). Such a discrepancy may be attributed to the fact that the amount of soot in this section is too low for the phase-lag to reflect in the global \(f_{\phi}^{\text{max}}\) response.

In summary, the dynamic response of soot to curvature fluctuations suggests that when the oscillation frequency \(f_{\phi}\) is increased, the soot formation time for bigger-sized particles tends to become larger than or comparable to the oscillation time. As a result, the larger-sized soot particles do not get enough time to respond to the imposed oscillations, which leads to their delayed response, causing a large phase shift in the soot response. In contrast, at lower frequencies, the characteristic time scales of larger-sized soot particles are smaller than the time scales of imposed oscillations. Therefore, the phase shift observed in the induced soot response tends to be smaller. It is also important to note that the flame curvature also impacts the characteristic time scales of soot formation for different sizes of soot particles, which further influences the dynamic response of soot formation to the imposed curvature oscillations.

### 4.1.2. Variation in the amplitude of imposed oscillations

In addition to frequency, the amplitude of the imposed curvature oscillations is expected to have a strong influence on the soot dynamics [12,15]. Therefore, to study the response of soot to the variation in amplitude at constant \(f_{\phi}\), additional computations have been performed with \(\Delta \kappa = 2\text{cm}^{-1}\) and \(8\text{cm}^{-1}\). Fig. 19 shows the dynamic response of \(f_{\phi}^{\text{max}}\) against time for different values of \(\Delta \kappa\).

From the numerical results, it is evident that the value of the induced amplitude of \(f_{\phi}^{\text{max}}\) depends strongly on the amplitude of the imposed oscillations in the flame curvature. The observed dynamic response of \(f_{\phi}^{\text{max}}\) at different amplitudes transitions from the strong increment in soot concentration under negative curvatures as noticed in Section 3.1. This is because, when the amplitude of imposed oscillation is increased, the flame encounters a larger range of negative curvature during the transient evolution. As a consequence, the soot mass growth rate is accelerated, which essentially reflects in the increased amplitude of the \(f_{\phi}^{\text{max}}\) response. Moreover, larger imposed amplitudes lead to a large asymmetry in the induced amplitudes with respect to the steady-state values. However, the phase-lag is hardly affected by the amplitude of the imposed curvature oscillations.

### 4.2. Dynamic response of soot to fluctuating curvature and fluctuating strain rate

During the steady flames analysis in Section 3.2, it was identified that the sensitivity of soot formation in the presence of flame curvature is greatly influenced by the strain rate variation. Therefore, it would be interesting to investigate the dynamic response
of soot formation to the simultaneously imposed unsteady fluctuations of strain rate and curvature.

4.2.1. Variation in strain rate oscillation frequency for the flat flame

Prior to studying the effects of simultaneous curvature and strain rate fluctuations, it is important to briefly highlight the dynamic behavior of soot for the flat flames (κ = 0) under the strain rate oscillations. Accordingly, the response of \( f_{\text{v}}^{\text{max}} \) (normalized by steady flat flame value) is compared against the time of imposed strain rate oscillations (normalized by the period \( \tau_a = f_a t \)) in Fig. 20 for \( \Delta \alpha = 50 \text{s}^{-1} \) at \( f_a = 50 \text{Hz} \). In Fig. 20 it can be observed that the induced oscillations of \( f_{\text{v}}^{\text{max}} \) are significantly damped and phase-lagged for an increase in the frequency of imposed strain rate oscillations. These observations are aligned with the findings reported elsewhere [12,13,15].

It is ratified in previous studies [12,39], that the strain rate oscillations mainly impact the thickness of the diffusion layer of the flame, and hence, influence the diffusion time of different species. Since a finite time is required for the species to be transported to the main reaction zone, the reaction zone essentially responds with a time lag when faster perturbations are introduced to the flow field by increasing oscillation frequency. Therefore, a larger phase-lag is expected at a higher frequency owing to the larger diffusion time of different species compared to the oscillation period. As a consequence, a hierarchical behavior between temperature, soot precursors, and soot sections is observed [15] regarding the phase-lag in their temporal responses. Furthermore, the overall soot response under strain rate fluctuations is found to be governed by the slow chemistry of larger-sized soot particles [15].

To demonstrate the validity of this correlation, the dynamic response of \( \omega_{\text{v}}^{\text{max}} \) is presented in Fig. 21 for different sections. It is evident that the phase-lag between imposed and induced oscillations is stronger at the higher sections (hence larger-sized soot particles). Therefore, similar to the case of curvature fluctuations discussed earlier, variability in the dynamic response of \( f_{\text{v}}^{\text{max}} \) can also be attributed to the transient response of \( \omega_{\text{v}}^{\text{max}} \) for different particle sizes owing to the large disparity in their time scales. However, it is interesting to note that the phase-lag observed in the \( \omega_{\text{v}}^{\text{max}} \) response at lower sections (e.g. \( i = 5, 10 \)) for pure strain rate fluctuations is slightly larger than the same noticed under pure curvature fluctuations. It further suggests that the soot response in terms of the phase-lag is more sensitive to strain rate fluctuations as compared to curvature fluctuations at the same frequencies.

4.2.2. Curvature and strain rate oscillations imposed at the same frequency

In this section, we compare the dynamic response of soot in flames subjected to simultaneous curvature and strain rate oscillations at identical frequencies. Accordingly, in Fig. 22, the response of \( f_{\text{v}}^{\text{max}} \) under simultaneous strain rate and curvature fluctuations
at $f_k = f_a = 50$Hz is compared against its response for the pure curvature fluctuations ($f_a = 0, f_k = 50$Hz) and pure strain rate fluctuations ($f_k = 0, f_a = 50$Hz). The dotted gray curve in Fig. 22 represents the expected $f_p^{\text{max}}$ after multiplying the effects of pure curvature fluctuations and pure strain rate fluctuations. Since the imposed oscillations of curvature and strain rate have identical frequencies (in-phase), the curvature and strain rate fluctuate simultaneously.

It is recognized from the analysis of steady flames (refer to Section 3.2) that under the decreasing strain rate, the presence of negative curvature tends to synergistically enhance the formation of soot. During the combined strain and curvature fluctuations, the flame encounters simultaneously decreasing strain rate and increasing negative curvature in every oscillation cycle. The flame with $f_k = f_a$, therefore, presents the local conditions favorable for soot formation during their transient evolution. As a result, the amplitude of $f_k^{\text{max}}$ fluctuations is found to be synergistically increased in the $f_k = f_a$ case compared to the $f_k = 0$ and $f_a = 0$ flames. Note that during the simultaneous strain and curvature oscillations, the flame is also subjected to increased strain rate and positive curvature, which tend to lower the soot formation. However, the sensitivity of soot formation suppression to positive curvature at higher strain rates was observed to be marginal (refer to Section 3.2). Therefore, the amplitude enhancement of $f_k^{\text{max}}$ response in the presence of simultaneously fluctuating strain and curvature shows strong asymmetric behavior, with the peak increasing faster than the reduction of the trough.

To understand the aforementioned observation better, the dynamic response of peak $\dot{\omega}^{\text{max}}$ for different sized particles is presented in Fig. 23. The profiles of expected $\dot{\omega}^{\text{max}}$ by multiplying the effects of $f_k = 0$ and $f_a = 0$ cases are plotted with the dotted gray curve. The $\dot{\omega}^{\text{max}}$ response under simultaneous strain rate and curvature fluctuations reveals that the synergistic effect of strain and curvature variation starts to appear for larger particle size. For instance, in the section $i = 30$, the amplitude enhancement under combined strain and curvature fluctuation is found to be higher than expected. As mentioned previously, the dynamic response of soot volume fraction is governed by the larger-sized soot particles, therefore, synergistic behavior noticed in soot mass growth rates for higher sections is also reflected in $f_p^{\text{max}}$ variation.

Note that, for larger-sized particles, the observed phase-shift in the $\dot{\omega}^{\text{max}}$ response under the presence of simultaneous strain and curvature fluctuations is substantially different than the one noticed in the algebraically predicted response (gray curve). As a consequence, the disparity in actual and algebraically predicted phase-shift in the $f_p^{\text{max}}$ response under simultaneously fluctuating strain and curvature is observed in Fig. 22. Furthermore, compared to the flames subjected to pure curvature and pure strain rate oscillations, the $\dot{\omega}^{\text{max}}$ response for the flame with simultaneously fluctuating strain and curvature tends to display a small phase-gain.

The dynamic response of $f_p^{\text{max}}$ under separate and simultaneous fluctuations of strain rate and curvature can be summarized in the combined plot of the unsteady limit cycle, as shown in Fig. 24. The solid curves in the figure correspond to the variation of $f_p^{\text{max}}$ with curvature (bottom abscissa) and strain rate (top abscissa) under steady conditions. The transient variation of $f_p^{\text{max}}$ against the strain rate for $f_k = 0$, and against curvature for $f_a = 0$ are compared with the unsteady limit cycle obtained under their simultaneous fluctuations at $f_a = f_k$. It is evident that, when curvature and strain rate oscillations are imposed simultaneously at the same frequency (in phase), the amplitude of induced oscillations in $f_p^{\text{max}}$ increases with a significant synergistic effect. Furthermore, for a given frequency, the response of soot tends to be more ‘decoupled’ from the imposed strain rate oscillations (indicated by the smaller amplitude of induced oscillations and large phase-lag) as compared to curvature oscillations. It further suggests that the cut-off frequency, beyond which the decoupling between soot chemistry and hydro-
dynamics occurs, would be higher for curvature than strain rate oscillations.

To highlight the impact of curvature and strain rate fluctuations on gas-phase quantities, the dynamic response of temperature and key chemical species is compared in Fig. 25. The induced oscillations of peak temperature hardly show any phase-lag for the pure strain rate and pure curvature fluctuations. For identical imposed frequencies, a phase-lag in the transient response of \( \text{C}_2\text{H}_2 \) for pure strain rate fluctuations is larger compared to the one for pure curvature fluctuations. It implies its stronger sensitivity to strain rate oscillations as compared to curvature oscillations. The observed sensitivity of gas-phase species to strain rate and curvature oscillations stems from the characteristic time scales governing their chemistry. As a result, for \( \text{H} \) radicals, which are characterized by fast time scales, the impact of curvature and strain rate oscillations appear quasi-steady. On the other hand, for \( \text{A4} \) owing to its slower characteristic time scales, the phase-lag can be noticed in the induced oscillations.

4.2.3. Curvature and strain rate oscillations imposed at non-identical frequencies

We further extend the analysis to compare the dynamic response of soot formation under unsteady curvature and strain rate fluctuations varying at different frequencies. For this analysis, two possible scenarios are considered based on the relative frequencies of curvature and strain rate oscillations. These include:

1. \( \kappa < f_a \): \( (\kappa = 50\,\text{Hz}, f_a = 100\,\text{Hz}) \)
2. \( \kappa > f_a \): \( (\kappa = 75\,\text{Hz}, f_a = 50\,\text{Hz}) \)

The dynamic response of \( f_V^\text{max} \) is compared in Fig. 26 for the aforementioned cases. In addition, the transient evolution of \( f_V^\text{max} \) for \( \kappa = f_a \) is also presented for reference. The numerical results demonstrate that, when curvature fluctuations are imposed in addition to the strain rate fluctuations, the overall soot formation process is strongly affected by the relative tendencies of curvature and strain rate to enhance or diminish the soot formation. For example, it is noticed that when the frequency of curvature and strain rate oscillations is identical \( (\kappa = f_a) \), the amplitude of the induced \( f_V^\text{max} \) fluctuations is maximum. On the other hand, for
the cases, in which the strain rate fluctuates at a frequency distinct from the frequency of curvature, multiple peaks are detected in the induced $f_{\max}^\alpha$ response.

For the case in which strain rate fluctuates faster than curvature ($f_\nu > f_\alpha$), close observation of $f_{\max}^\nu$ response and imposed curvature-strain rate oscillations indicate that the first maximum in the $f_{\max}^\nu$ response occurs when the flame is subjected to a lower strain rate along with decreasing positive curvature, while the second maximum occurs when both strain rate and curvature tend to reduce, with the latter being negative. Since for the first peak, the curvature is positive, the $f_{\max}^\nu$ value for the first peak is found to be smaller than the one observed for the second peak. Furthermore, a higher strain rate frequency tends to enhance the phase-lag and damping of $f_{\max}^\nu$ amplitude as compared to its response under in-phase conditions ($f_\nu = f_\alpha$).

On the other hand, when the frequency of the imposed curvature oscillations is higher than that of the strain rate fluctuations ($f_\nu < f_\alpha$), the number of extrema in the $f_{\max}^\nu$ response increases. This is because when the frequency of curvature oscillation is increased, the number of curvature cycles per strain rate cycle is enlarged. As a result, the flame is subjected to multiple conditions that either assist or impede the rates of soot formation. Accordingly, for the $(f_\nu > f_\alpha)$ case considered here, three maxima are noticed in the $f_{\max}^\nu$ evolution. These peaks in $f_{\max}^\nu$ arise, respectively, as a consequence of the following instances: (i) when both strain and curvature tend to decrease, with the strain rate reduction being slower than the curvature, while curvature being on the positive side. (ii) when the strain rate is on the low side and curvature tends to decrease from positive towards negative. (iii) when both strain and curvature decrease with the strain rate drop being faster than the curvature decrement. The maximum peak observed in the overall $f_{\max}^\nu$ response is mainly the reflection of the last condition.

Note that the aforementioned observation is specific to the chosen values of $f_\nu$ and $f_\alpha$. In a general sense, the dynamic response of $f_{\max}^\nu$ under simultaneous strain rate and curvature fluctuations indicate an additive effect of separately imposed strain rate and curvature oscillations. In other words, during the transient evolution, soot formation (indicated by $f_{\max}^\nu$) is found to be enhanced when a flame encounters local conditions favorable for soot formation (e.g. negative curvature at a low strain rate), while the soot formation is found to be reduced for the adverse local conditions (e.g. positive curvature at a high strain rate). The competition between local strain rate and curvature essentially determines the number of extrema (and their amplitudes) in the induced soot response.

From numerical results, it can be argued that, under simultaneously fluctuating curvature and strain rate, the variability in values of the induced $f_{\max}^\nu$ amplitudes strongly depends on the frequency difference between the imposed oscillations. In principle, the amplitude of induced soot response is found to be largest when imposed curvature and strain rate frequencies are identical (in-phase). On the contrary, the amplitude of induced oscillations attains the lowest value for the curvature and strain rate oscillations being in anti-phase (not shown here). For the frequency of curvature oscillations distinct from that of strain rate oscillations, the number of extrema and their corresponding values in induced soot response are governed by the magnitude and balance between the imposed frequencies.

5. Concluding remarks

The response of soot formation to curvature in an SF type counterflow flame was numerically investigated under steady and unsteady conditions using detailed kinetics and a discrete sectional method-based soot model. A detailed analysis of various soot sub-processes was conducted to acquire insights into the associated mechanism. The analysis was extended to study the sensitivity of soot formation in curved flames to strain rate variation. The dynamic response of soot to externally imposed fluctuating curvature was successively studied in a wide range of conditions. Furthermore, simulations were conducted to investigate the competing effects of simultaneously imposed strain rate and curvature fluctuations on the transient response of soot formation.

The present numerical study revealed some interesting findings which can be summarized as follows:

1. The numerical results for steady counterflow flames showed that in presence of curvature, due to a change in the surface area along the flame, the velocity of soot particles is significantly altered. For negatively curved (center of curvature lies in the fuel-rich side of the flame front) flames, the soot particle velocity is found to be lowered compared to flat flame. The lowered soot velocity leads to an enhanced residence time in the soot formation zone and consequently contributes to higher volume fractions. Furthermore, the flame curvature strongly affects the differential transport of soot particles with respect to the gas phase, due to their high Schmidt numbers. For negative curvature, soot particles are differentially transported towards the lower mixture fractions, leading to their accumulation within the soot formation zone (characterized by region in Z-space with large rates of soot sub-processes). Soot displaced towards the soot formation zone grows predominantly by surface reactions leading to increased soot mass and volume fractions. Subsequent to surface growth, the curvature-induced transport of large-sized particles also promotes their coagulation which further contributes to the increase in the soot surface area available for surface growth reactions. Positive curvature on the other hand exhibits the opposite behavior causing lowered soot concentration. The role of PAH in overall soot response under curvature is found to be secondary.

2. Negative curvature leads to a synergistic increment in soot volume fractions at lower strain rates compared to their flat flame counterparts, indicating strong complementary effects of strain rate and curvature. On the contrary, in positively curved flames, such synergistic effects are hardly exhibited.

3. The imposed curvature oscillations tend to increase the soot concentration compared to steady-state conditions. Curvature fluctuations significantly impact the distribution of soot within the flame. At lower frequencies the response of soot to curvature oscillations is quasi-steady. At intermediate oscillation frequencies, the dynamic response of soot is phase-lagged, and the phase-shift in the induced oscillations tends to increase with frequency. The overall response of soot volume fraction is recognized to be predominantly governed by the dynamics of larger-sized particles owing to their large characteristic time scales. With an increase in the imposed frequency, the dynamic response of soot gets attenuated leading to the lowered amplitudes of the induced oscillations. When the frequency of imposed fluctuations is sufficiently large, soot particles do not adequately respond to the faster curvature fluctuations, and the soot response becomes decoupled from curvature.

4. A more complex flow-flame-soot interaction is observed when the flame is subjected to simultaneous strain rate and curvature fluctuations. For in-phase strain rate-curvature oscillations, the soot formation is enhanced due to the complementary effects of strain and curvature. At unequal frequencies, on the other hand, the locally adverse/favorable conditions encountered by the soot formation process during transient evolution govern the overall dynamic response of the soot.

In conclusion, the present work demonstrates that the soot response in curved counterflow flames is strongly correlated to the
residence time of soot particles in the soot formation zone and differential diffusion effects. The discrete sectional method-based model provides important insights into the curvature effects on the soot particle size distribution. Furthermore, the sectional model facilitates the investigation of the damped and delayed response of soot to oscillating curvature in correlation with the variability in the response of different sized soot particles.

Given the strong impact of curvature on soot formation, combustion models relying on a one-dimensional description of flat flames may not be adequate in predicting the soot characteristics in strongly curved flames. Therefore, integration of curvature effects in flamelet-based models would be instrumental, and in combination with a detailed soot model, they might deliver accurate soot results in multi-dimensional sooting flames.

It is important to recognize that, although the retained soot modeling approach is state-of-the-art, due to the lack of reliable experimental validation, it may still present certain undiscovered limitations in capturing physics related to curvature effects in its entirety, and thus, to establish the general consensus about the presented conclusions. Therefore, the analysis concerning the role of different soot sub-processes in curvature effects may depend on the employed soot model in conjunction with the gas-phase kinetics.

Moreover, it is worth highlighting that although a simplified 1D analysis was conducted to study curvature unsteadiness, the chosen frequency modes or curvature values in the counterflow setup were not explicitly analyzed concerning the turbulent combustion regime. Hence, a direct correlation of this laminar flame study to turbulent combustion is unexplored at the moment, and further fundamental studies are therefore still necessary. Nevertheless, the analysis of unsteady curvature effects on soot can be recognized as a step forward for a better understanding of the interactions between soot kinetics and fluid dynamics to guide further modeling efforts.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Acknowledgements

The research leading to these results has received funding from the European Union’s Horizon 2020 Programme under the ESTi-Mate project, grant agreement No. 821418.

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