

Frequency response of counter flow diffusion flames to strain rate harmonic oscillations

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The formation of pollutant species in turbulent diffusion flames is strongly affected by turbulence/chemistry interactions. Unsteady counter flow diffusion flames can be conveniently used to address the unsteady effects of hydrodynamics on the pollutant chemistry, because they exhibit a larger range of combustion conditions than those observed in steady flames. In this paper, unsteady effects on the formation of PAH and soot are investigated by imposing harmonic oscillations in the strain rate in a large range of frequencies and amplitudes.

PAH and soot exhibit specific behaviour according to their characteristic chemical times which are longer than those of the main combustion process. At low frequencies of imposed oscillations the PAH and soot profiles show strong deviations from the steady-state profile. At large frequencies a decoupling between the PAH and soot concentration and the velocity field is evident. Soot formation is found less sensitive to velocity fluctuations for flames with large initial strain rate. Numerical results are successfully compared with experimental measurements obtained in similar conditions.

1. Introduction

Turbulent non premixed flames are largely used in many practical combustion devices which need to respect always more stringent limitations on pollutants emissions. Turbulent flames are difficult to study, due to the complex interactions and the coupling between spatial and time scales of fluid dynamics and chemistry. Moreover, the formation of pollutant species can be accurately predicted only if large and detailed kinetic schemes are used. The Steady Laminar Flamelet Model (SLFM) represents one of the most used approaches to numerically simulate complex turbulent non premixed flames (Peters, 1986; Peters 2000). The flamelet approach implicitly assumes that the flamelets respond in a quasi-steady manner to the local strain rate variations. However, in a real combustion device, the strain rate can fluctuate around its main value (which is established by the large scale eddies) because of the smaller eddies with characteristic turnover times comparable to the characteristic diffusion times. If Reynolds number is sufficiently high, a large range of eddy sizes is expected, in which the characteristic eddy turn-over times become comparable with the diffusion time in the laminar flamelet (Im *et al.*, 2000). In such conditions, the unsteadiness begins to show its effects on the characteristics of the laminar flamelets and consequently it becomes important to understand the unsteady effects and if information obtained in steady state conditions can be extended to unsteady conditions (Haworth *et al.*, 1988; Barlow *et al.*, 1992; Egolfopoulos *et al.*, 1996; Xiao *et al.*, 2005). From a numerical point of view the

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introduction of unsteadiness in combustion is a challenge because of the strong coupling between the time and spatial scales of convection, diffusion and reaction. As a first step, many authors suggested to perform this kind of investigations by exposing the laminar flames to far-field harmonic oscillations in the strain rate, using a large range of frequencies (Im *et al.*, 2000; Egolfopoulos *et al.*, 1996; Decroix *et al.*, 1999; Decroix *et al.*, 2000). The effects of unsteadiness on counterflow diffusion flames were experimentally (Xiao *et al.*, 2005; Decroix *et al.*, 1999; Decroix *et al.*, 2000, Welle *et al.*, 2000) and numerically (Im *et al.*, 2000; Egolfopoulos *et al.*, 1996, Stahl *et al.*, 1991; Cuoci *et al.*, 2008) investigated by several authors. From both experimental and numerical results, it seems evident that the assumption of quasi-steady response in some conditions is not appropriate. In particular at low or intermediate values of the oscillation frequency the behavior of the flame response is very complex, especially for pollutant species, such as Polycyclic Aromatic Hydrocarbons (PAH) and soot, whose chemistry is slow.

In this paper the transport equations governing the dynamic behavior of the flame are numerically solved using a detailed kinetic scheme and an accurate description of the transport properties. Harmonic oscillations of the steady values of the fuel and oxidizer stream velocities are externally imposed and affect the flame strain rate.

2. Numerical Method

The counter flow configuration investigated in the present work consists of two nozzles which produce an axisymmetric flow field with a stagnation plane between the nozzles. The two-dimensional flow is reduced mathematically to one dimension by assuming that the radial velocity varies linearly in the radial direction, which leads to a simplification in which the fluid properties are functions of the axial distance only. The numerical solution of the flames in this counter flow configuration is obtained by solving the unsteady conservation equations of mass, momentum, energy and species concentrations (Cuoci *et al.*, 2008). Initially the problem for the steady flame is solved: the corresponding solution is assumed to be the starting point for the application of the harmonic oscillations (Egolfopoulos *et al.*, 1996). It is assumed that the oscillations of the inlet velocities have the following form:

$$u(t) = u_{steady} [1 + A_u \cdot \sin(2\pi f \cdot t)] \quad (1)$$

3. Kinetic Scheme

Detailed kinetic models can be effectively used to analyze the combustion of hydrocarbons and the formation of pollutant species such as PAH and soot. For this purpose, in addition to pyrolysis and oxidation reactions that convert and oxidize hydrocarbons, it is also necessary to include several classes of condensation and dealkylation reactions that govern the growth of PAH and soot. Moreover, the large number of reactions that are required to describe the combustion of large hydrocarbons precludes the possibility to use a fully detailed approach to describe their combustion mechanism. A semi-detailed or lumped approach can be conveniently adopted to reduce the overall complexity of the resulting chemical-kinetic scheme, both in terms of

equivalent species and lumped or equivalent reactions (Granata *et al.*, 2003, Ranzi *et al.*, 2007). The extension to the modelling of soot formation can refer to the discrete sectional method. PAHs larger than 20 C atoms and soot particles are divided into a limited number of classes covering certain mass ranges. Each class is represented by two or three lumped pseudo-species, called BIN, with a given number of carbon and hydrogen atoms. The resulting kinetic scheme involves 253 species in ~14,000 reactions. The general features of this approach were recently presented and discussed (Ranzi *et al.*, 2005; Granata *et al.*, 2006). The thermo-chemical data are obtained primarily from the CHEMKIN thermodynamic database (Kee *et al.*, 1989).

4. Unsteady Combustion

4.1 Oscillating Perfectly Stirred Reactor

In order to better understand the turbulence/chemistry interactions, a Perfectly Stirred Reactor (PSR) can be conveniently considered. The effects of turbulent fluctuations can be “*simulated*” by forcing the reactor temperature to oscillate sinusoidally around the mean value T_{av} , while the residence time is kept fixed. Temperature oscillations induce oscillations in the composition. In particular, the responses of specific chemical species are expected to be different, according to the characteristic chemical times.

Figure 1 shows the concentrations of PAHs and soot as a function of the non dimensional time (ratio of time and oscillation period). PAHs and soot are strongly affected by the temperature oscillations: their profiles are highly asymmetric and do not follow the sinusoidal shape. Since the reactions governing the formation of chemical species are non linear, a difference exists between the mean value of the oscillating composition and the value corresponding to the mean temperature of the reactor. Figure 2 compares the mean and the steady state values of PAHs and soot. The behavior is quite complex. PAHs shows a maximum at ~1600K when steady state conditions are assumed, while the value obtained by averaging the oscillations is smaller than the steady state value in the range ~1400-1700K and larger at higher temperatures. Even more complex is the result for soot: the mean values (which account for temperature oscillations) do not show the maximum at ~1700 K.

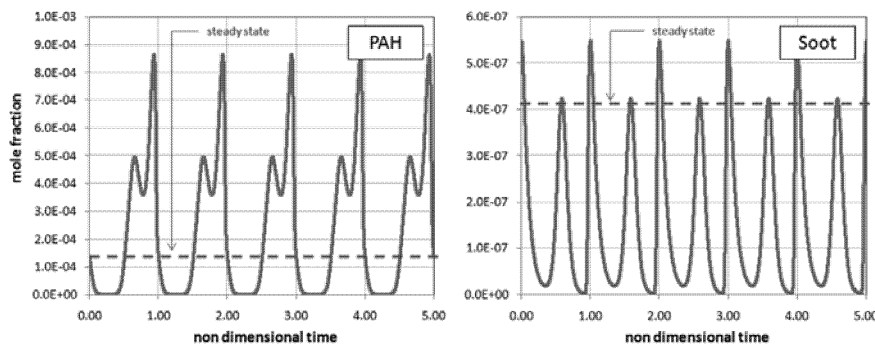


Figure 1. Profiles of PAH and soot in an oscillating PSR. Mixture of propane/air, $\Phi=2$. $T_{av}=1800K$, $\Delta T=360K$, $f=100$ Hz.

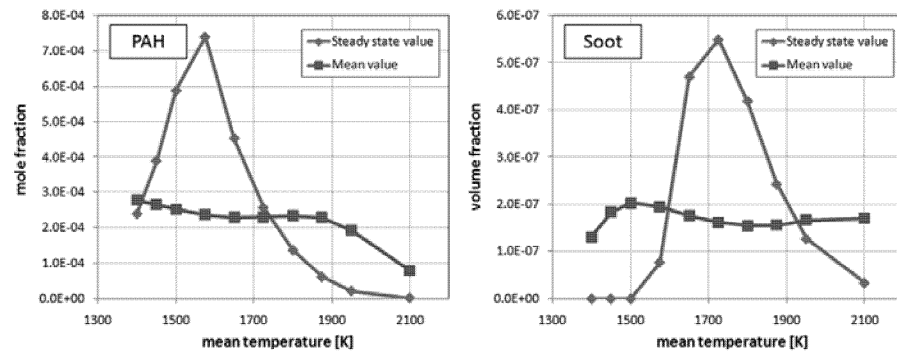


Figure 2. Comparison between steady state and mean value of PAH and soot in the oscillating PSR. The operating conditions are the same of Figure 1.

4.2 PAH Formation in Oscillating Counterflow Diffusion Flames

The counter flow diffusion flames numerically simulated in this paper were experimentally investigated by (Xiao *et al.*, 2005). The oxidizer and the fuel tubes are 25.4 mm in diameter and have a 12.7 mm separation distance; the inlet temperatures are 298K. Three different strain rates were studied as briefly described in Table 1.

Table 1. Operating conditions for the three flames investigated

	Flame I	Flame II	Flame III
Global Strain Rate [Hz]	23	44	74
Fuel Velocity [cm/s]	25.71	50.11	75.82
Air Velocity [cm/s]	18.92	38.10	57.25

The steady-state analysis of the structure of these flames is already described in detail in (Cuoci *et al.*, 2008). These calculations are used as a starting point for the application of a forced harmonic oscillation on the inlet velocities using the same amplitude and phase. The position of the stagnation plane, which depends on the stream momenta, oscillates accordingly. In Figure 3 the numerical results obtained for Flame II are summarized in terms of temperature and PAH mole fraction peak values. Following (Xiao *et al.*, 2005), PAHs were grouped in three classes. Class A includes one- and two-rings PAHs, Class B three- and four-rings PAHs and Class C PAHs with more than four rings.

Panel a of Figure 3 shows the variation of the maximum temperature induced by the oscillation of velocities of the streams. It is evident that the amplitude of the oscillations decreases at high frequencies. On the contrary the phase lag, which tends to zero for low values of the frequencies, becomes larger. When a low frequency is used, the induced oscillation amplitude of the PAH profiles is very large and the symmetry about the mean value is lost. However the phase lag is small, which indicates a quasi-steady response of the smallest PAHs in these conditions. For Class B and Class C the same observations can be repeated (Panels c and d of Figure 3). Moreover the maximum frequency for which the flame response can be considered quasi-steady for larger PAHs. These results are in agreement with experimental and numerical investigations performed by several authors (Egolfopoulos *et al.*, 1996; Xiao *et al.*, 2005). The non-

dimensional Stokes' number (η_K) can be useful to better understand the frequency response of counter flow diffusion flames:

$$\eta_K = \sqrt{\frac{\pi \cdot f}{K}} \quad (2)$$

where f is the frequency and K is the cycle mean strain rate.

The Stokes' parameter compares the characteristic time of imposed oscillations, ($1/f$), with the characteristic time of diffusion phenomena in the flame, which can be approximated as $\sim 1/K$. When the Stokes' parameter is low the diffusion time is lower than oscillation time and therefore a small phase-lag is expected between the imposed and induced oscillations. At higher η_K values large phase shifts can be observed in the flame behavior. Figure 4 shows the peak value of the mole fraction of each PAH class versus the Stokes' number. The numerical results are compared with the experimental measurements obtained in the same conditions by (Xiao *et al.*, 2005). Experimental and numerical results agree and confirm the large amplitudes of oscillations for Class C, especially for low Stokes' numbers.

As expected the ratio between the peak and the steady state values are larger when the Stokes number is small. The amplitude of the oscillations is strongly damped for large Stokes numbers because of the corresponding high values of oscillation frequency.

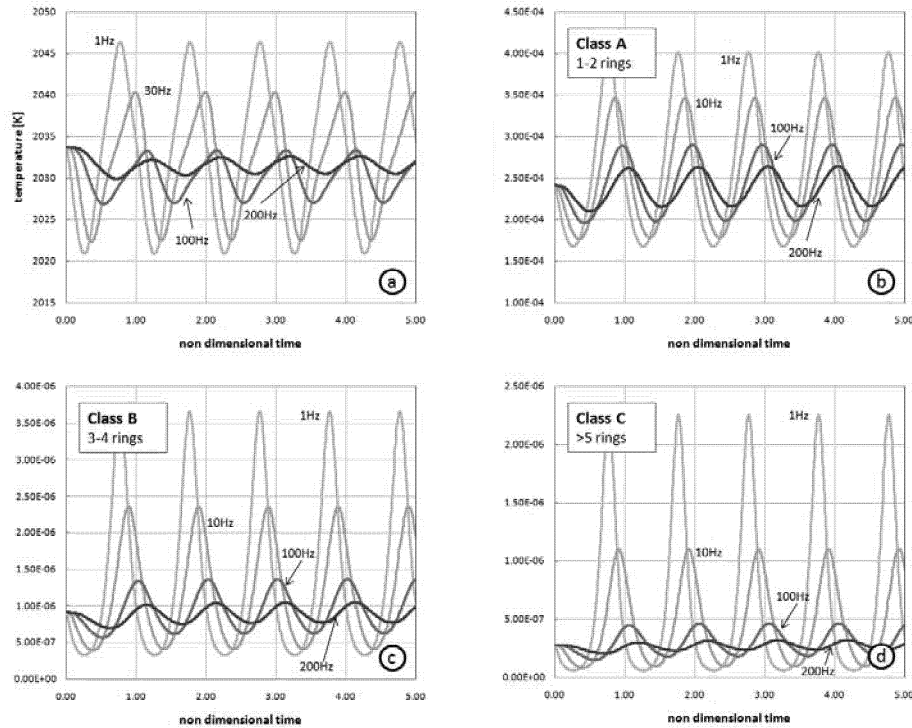


Figure 3. Flame II: maximum flame temperature and peak values for each PAH class at different frequencies, non dimensional time $t' = \text{time}/\text{period}$ ($A_u = 0.5$).

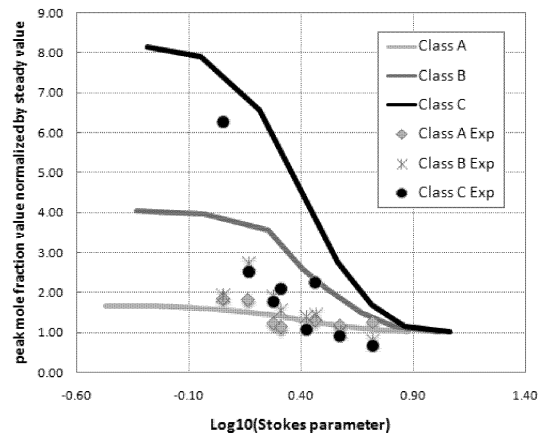


Figure 4. Normalized mole fraction peak values versus the Stokes number compared with experimental measurements (Xiao *et al.*, 2005) (Flame II, $A_u=0.7$ of the steady state value). The profiles are scaled by the steady state value.

4.3 Soot Formation in Oscillating Counterflow Diffusion Flames

The formation of soot was simulated on the counterflow diffusion flames experimentally investigated by (Decroix *et al.*, 2000) and partially by (Xiao *et al.*, 2005). Three different fuels (CH_4 , C_3H_8 , C_2H_4) were investigated at four different global strain rates (GSR) ranging from 15, to 90 Hz.

The steady-state analysis of the structure of these flames is already described in detail in (Cuoci *et al.*, 2008b). Only some details are discussed here. The flames are located on the oxidizer side of the stagnation plane, therefore oxygen is supplied to the flame by both convection and diffusion, while the fuel is supplied mainly by diffusion. The fuel is completely decomposed before reaching the flame front, while oxygen is able to penetrate deeply in the fuel zone. After inception on the fuel side, soot particles are convected away from the flame toward the stagnation plane. As a consequence soot oxidation is absent. Two diffusive layers (fuel and oxidizer) can be observed, whose thickness is inversely proportional to the strain rate. These diffusive layers are the main responsible of the characteristics of the flame response to the imposed oscillations. Figure 5a compares the normalized axial profiles of OH and soot of a propane-air flame with measurements (Decroix *et al.*, 2000). The numerical results correctly predict the location of the flame front (OH profile) and of the soot region. Figure 5b shows the good agreement between the predicted and measured maximum soot volume fraction as a function of the global strain rate. The soot volume fraction decreases with the strain rate, which is inversely proportional to the characteristic residence time. As evident, the soot volume fraction is a strong function of the fuel type (C/H ratio) and is larger for ethylene. These steady-state calculations are used as a starting point for the application of a forced harmonic oscillation of inlet velocities using the same amplitude and phase. Figure 6 shows the detailed frequency response of soot to the imposed oscillations. The steady state value, the peak and through values and the mean value (which accounts for the oscillations) of soot are reported as a function of the externally imposed frequency together with the phase shift. The amplitude of the oscillations is largely reduced at

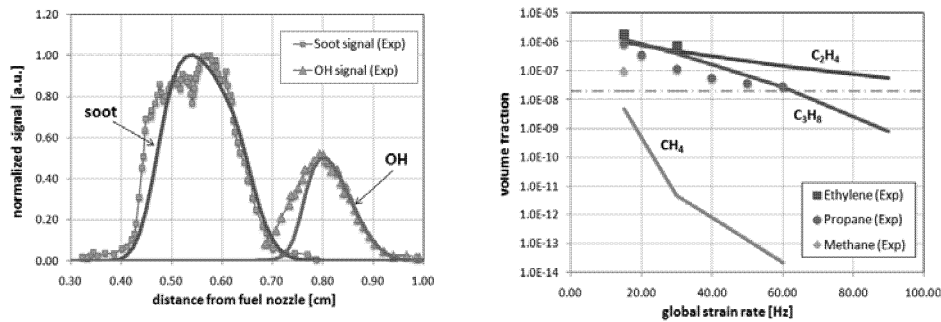


Figure 5. (a) Comparison between predicted and experimental (Decroix et al., 2000) profiles of OH and soot (propane flame, GSR=15 Hz). (b) Peak soot volume fraction. The lines indicate predictions, symbols measurements. The dotted line indicates the minimum experimental amount of detectable soot.

higher frequencies, while at low frequencies the flame response to the changes in the inlet velocity value can be considered quasi-steady.

A non symmetrical behavior is particularly evident at low frequencies. The main effect of oscillations is an increase in soot production. When the frequency is large the mean values tend to approach the steady value (and therefore the unsteady effects are less important), but the flame response cannot be considered quasi-steady because of the large phase-lag. The flame response can be explained by comparing the diffusion time in the flame structure with the oscillation time. At low frequencies the diffusion time is small and therefore the response is quasi-instantaneous. At higher frequencies the diffusion time becomes larger than the oscillation time and therefore a phase lag arises between the imposed and induced oscillations. The overall response of the flame can be very complex because the diffusion time is different for every chemical species and therefore each species responds in a different way.

Due to the phase lag, the temperature field is not in phase with the velocity field: for a quasi-instantaneous response, the minimum temperature should correspond to the maximum velocity location (minimum residence time). On the contrary, the predicted

Phase lag	Exp.	Num.
Max velocity		
÷	~90°	~87°
Min temp.		
Max velocity		
÷	~125°	~118°
Min Soot		
Max PAH		
÷	~270°	~290°
Max Soot		

Table 2. Measured (Santoianni et al., 2001) and predicted phase lag (C_3H_8 flame, $A_u=0.6$, $f=25$ Hz)

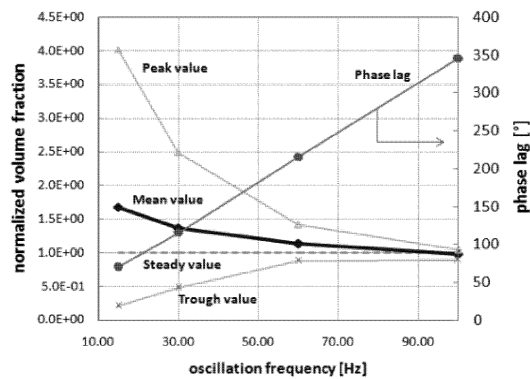


Figure 6. Detailed frequency response of the oscillation amplitude and phase lag of soot (propane flame, GSR=60 Hz, $A_u=60\%$).

minimum temperature occurs with a phase shift of $\sim 87^\circ$. Moreover, the minimum soot volume fraction should occur at the maximum velocity: the calculated phase shift between the minimum of soot and maximum velocity is $\sim 118^\circ$, while the phase lag between the maximum PAH and maximum soot is $\sim 290^\circ$. These numerical results are in good agreement with the experimental measurements performed by (Santoianni *et al.*, 2001) on the same flame as summarized in Table 2.

5. Conclusions

The effect of strain rate fluctuations on the formation of PAH and soot was numerically investigated in unsteady counterflow diffusion flames using a very detailed kinetic scheme. Several counterflow diffusion flames were studied and the numerical results confirmed the experimental observations: the oscillations tend to increase PAH and soot production with respect to steady-state conditions. The response of the flame is complex and depends on the frequency of the imposed oscillations, the initial strain rate and the characteristic chemical times of the different species: when the imposed frequency is large, PAH and particularly soot do not appreciably respond to the fast strain rate fluctuations. On the contrary, sinusoidal responses can be observed for low or intermediate oscillation frequencies in the instantaneous strain rate.

6. References

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