The effect of fine droplets on laminar propagation speed of a strained acetone-methane flame: experiment and simulations

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Abstract

In this study, we investigate the effect of the presence of fuel droplets, their size and concentration, on stretched laminar flame speeds. We consider premixed strained methane/air mixtures, with the addition of small acetone droplets, and compare the flame velocity field behavior to that of the fully vaporized mixture. An impinging stagnation flame configuration is used, to which a narrowly distributed polydisperse mist of acetone droplets is added. Total acetone molar concentrations between 9\% and 20\% per mole of methane are used, corresponding to 18.6\% and 41.4\% of the total fuel energy. The Sauter Mean Diameter (SMD) of acetone droplets is varied from 1.0 to 4.7 \(\mu\)m by carefully tuning the air flow rate passing through an atomizer. The

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droplet size distribution is characterized by a Phase Doppler Anamometry (PDA) system at the outlet of the burner. The flame propagation speed is measured using Particle Image Velocimetry (PIV) for overall equivalence ratios ranging from 0.8 to 1.4 at various strain rates, and the result is compared with a reference case in which acetone was fully vaporized. Unlike the fully vaporized flame, a two-stage reaction flame structure is observed for all droplet cases: a blue premixed flame front followed by a reddish luminous zone. Comparison of the results between gas-only and droplet-laden cases shows that the mean reference burning velocity of the mixture is significantly enhanced when droplets are present under rich cases, whereas the opposite is true for stoichiometric and lean cases. The mean droplet size also changes the relationship between flame speed and strain rate, especially for rich cases. The result suggests that with typical conditions found in laminar strained flames, even for the finest droplets that may have been vaporized before reaching the flame front, the resulting inhomogeneities may lead the flame to behave differently from the well-premixed gaseous counterpart. Simulations at similar conditions are performed using a two-phase counterflow flame model to compare with experimental data. Model results of reference velocities do not compare well with observations, and the possible reasons for this behaviour are discussed, including the difficulties in determining the pre-vaporization process and thus the boundary conditions, as well as the fidelity of the current point-source based 1D model.

Keywords: aerosol, sprays, laminar flame, PIV, flame speed

1. Introduction

Spray combustion has received considerable attention due to its wide application in internal combustion engines, gas turbines, and other energy conversion devices. Compared with gas-phase combustion, it is a significantly more complicated phe-
nomenon involving mixtures of gaseous and liquid fuels. Given their complexity, studies on spray combustion have grown into several sub-areas from droplet production, collision, evaporation, and finally, reaction. A number of comprehensive reviews have been produced on these topics [1–4]. In the present study, we mainly focus on the behavior of narrowly dispersed fuel droplets in strained laminar flames with controlled droplet size and concentration. The topic is directly relevant to the understanding of the flamelet formulation for spray flames, as discussed by various researchers [5–7].

Among the earliest experimental studies on the issue, Hayashi et al. [8] measured the laminar flame speed of stagnant droplet-vapor-air mixtures of ethanol and n-octane droplets in a combustion chamber, and concluded that the multi-phase mixture could burn faster than the gas-phase mixture, provided that the fuel fraction in the liquid phase remained small; for larger fractions, the flame was quenched by heat transfer to the fuel droplets. The experimental work by Nomura and co-workers [9] investigated the droplet size effect on flame propagation using a similar method. They reported that there was an optimal ethanol droplet size which maximized the burning velocity of the droplet-vapor-air mixture in a combustion chamber, and that this optimal size changed with overall equivalence ratio. Nunome et al. [10] observed unsteady flame propagation in n-decane sprays under microgravity conditions and found that the mean interdroplet spacing also had strong effects on the flame spread rate. A major disadvantage of using a combustion chamber, however, is the transient nature of the phenomenon, whereby it becomes more difficult to apply advanced laser diagnostics to probe local conditions. A second issue arises with ignition of the droplet-laden mixture, which must take place immediately after filling the chamber, otherwise deposition of large droplets due to gravity may affect data quality. To overcome these problems, several studies [11–13] introduced an opposed-jet (counterflow)
configuration, to take advantage of the well defined boundary layer conditions and ease of simulation. These counterflow experiments used a diffusion flame configuration in order to stabilize the flame, with droplets and carrier gas emerging from one port, and preheated oxidizer delivered through the other. Chen et al. [12] observed two very different flame structures among these studies: for low strain rates and small droplet sizes, the flame appeared as a thin blue sheet, whilst for high strain rates and large droplet sizes the flame exhibited an additional thick orange region on the oxidizer side, which indicated possible soot formation in this region. However, in the two cases studied in [12], droplet size, initial momentum and strain rate were coupled together, so a clear explanation of what caused this complex flame structure could not be given. Similar two-layer flame structures (thin blue sheet followed by a thick orange zone) were also observed in premixed spray flames on a stagnation burner, but no burning rate measurements were made [14, 15].

Conducting well-controlled experiments on laminar droplet-laden flames is challenging because: (a) methods to generate mono-dispersed fuel droplets are limited, and the droplet size distribution is often coupled to the liquid fuel concentration, (b) droplets experience a long evaporation time before reaching the reaction zone, making it difficult to know the accurate mass fraction of vapor and of droplet in the mixture, and (c) spray/air flames often cannot be sustained in the laboratory at the same condition as gaseous flames. Therefore, often pilot flames or carrier gas preheating are used to aid the stabilization, both of which may affect the flame structure. To the best of our knowledge, there have been no measurements of the burning velocity in droplet-laden premixed flames on steady laminar burners, and this is the primary motivation for the present work.

Several computational studies on droplet combustion have also attempted to address the issue of how droplets affect the propagation of premixed flames. One
example is the work by Neophytou and Mastorakos [16], where free propagation of a one-dimensional laminar flame through $n$-heptane or $n$-decane mixtures in air as point source droplets was simulated with detailed chemistry, and point-source formulation. In that numerical work, the enhancement of flame propagation speed was predicted for rich conditions, and an optimal droplet size was suggested, which maximized the burning velocity of the mixture. The optimal droplet size was related to the equivalence ratio, fuel volatility and residence time prior to entering the flame zone. Perhaps unsurprisingly, the maximum flame speed usually appeared under conditions where an effective equivalence ratio $\phi_e$, expressing the actual amount of vaporized fuel reacting at the flame front, was near unity. Apart from the free flame propagation model, a droplet combustion model based on the counterflow configuration has also been developed by Continillo and Sirignano [17], using a hybrid Eulerian-Lagrangian formulation, representing the droplets as a point source, and single-step chemistry. The two-dimensional counterflow was converted to a one-dimensional problem by conducting similarity analysis. That work suggested that for a premixed, droplet-laden flame in the counterflow configuration, a second reaction zone with diffusion-like characteristics might be observed after the premixed flame zone. Larger droplet sizes were found to cause a separation of the flame into a premixed flame zone and a diffusion flame, whereas an increase in strain rate suppressed this separation. Later on, Gutheil and Sirignano [18] added detailed chemistry to the counterflow flame model for methanol/$n$-heptane droplets in a diffusion configuration, and showed that two reaction zones were again observed for large droplets with high initial momentum, as they cannot be fully vaporized at the flame front, but instead penetrate the first reaction zone and form a second one. Their work also predicted that the flame temperature should peak at a certain level of strain rate and that further increases in the strain rate suppressed the flame, leading to extinction.
Lentati and Chelliah [19] developed a similar one-dimensional model which accounted for the dynamics of 5-50 µm water droplets in a counterflow non-premixed field with a simplified reaction mechanism to investigate flame extinction characteristics. A similar model was adopted by Kee et al. [20] to investigate the effects of thermophysical properties, carrier gases, and pressure on the reaction of fuel droplets using a diffusion counterflow configuration. Following the work of Gutheil and Sirignano [5, 18], Luo et al. [6] and Franzelli et al. [7] investigated methods of incorporating the presence of droplets in spray flamelet models, identifying the role of evaporation and species diffusion in mixture fraction space, and obtained limiting results for non-premixed strained flames. The only numerical work that investigated multi-step chemical kinetics in premixed counterflow droplet-laden flames is the work by Chen et al. [21], in which water droplets were added to a methane/air flame to predict the extinction characteristics of the flames. Premixed droplet-laden flame models are particularly of interest because the flame speed, one of the key parameters in the combustion theory and applications, is only defined in premixed flames.

The range of numerical studies on the fundamental mechanism of droplet-flame interactions has outpaced their experimental validation, owing to the difficulties in conducting well controlled experiments as described above. In particular, stabilizing a droplet-laden flame with a single fuel over a range of equivalence ratios is difficult for premixed flames. However, an alternative can be found by creating a hybrid binary mixture consisting of both gaseous and liquid fuel, which is more easily stabilized. In previous work [22], we measured the laminar flame speed of acetone/methane/air mixtures for a canonical impinging flame configuration, over a range of stoichiometries and mixture ratios, in which acetone was fully vaporized. The laminar flame speed was measured based on the stagnation flame method [22, 24]. In this method, the stretched flame speed is obtained from the minimum axial velocity upstream of
the flame front, and the local strain rate is extracted from the radial velocity profile at the reference plane. The unstrained flame speed can then be extrapolated from the corresponding strained values. The original data obtained in that study provides a baseline case to investigate how the presence of droplets may affect flame propagation. In the present work, we use the same technique to measure the stretched laminar flame speed of acetone mist/methane/air mixture for the same conditions as in the original study, except that the acetone fraction is now presented in both liquid and vapor phase (from pre-vaporization). The flame speed measurements as a function of strain rate and range of global equivalence ratios are presented, along with the reference flame speeds for different droplet sizes and strain rates.

Several important considerations arise with the presence of droplets when using the stagnation flame method, namely: (a) how much droplet vaporization takes place across the preheat and reaction zone, and what effect vaporization is on the surrounding temperature and mixture fraction, (b) differences between the droplet and surrounding gas velocity, and (c) whether the droplets produce any group effect over the tested concentration range. These issues are briefly discussed, as follows.

1.1. Effect of droplet vaporization

The criterion for vaporization depends on the ratio of the time for vaporization relative to the convection time into the flame. The time for vaporization is given as

\[ \tau_v = \frac{d^2}{8D \ln(1 + B_M)} \frac{\rho_t}{\rho_g} \]  

(1)
where $d$ is the droplet diameter, $D$ is the diffusivity, $\rho_l$ and $\rho_g$ are respectively the densities of the liquid and gas, and $B_M$ is the Spalding mass transfer number,

$$B_M = \frac{Y_{f,s} - Y_{f,\infty}}{1 - Y_{f,s}}$$  \hspace{1cm} (2)$$

where $Y_{f,s}$ is the mass concentration of fuel at the surface of the droplet, $Y_{f,\infty}$ correspondingly in the far field. The convective time in a stagnating flow is approximately given as the inverse strain rate $K$, so that the non-dimensional evaporation number is given as

$$Ev = \tau_v K = \frac{d^2 K}{8D \ln (1 + B_M)} \frac{\rho_l}{\rho_g}$$  \hspace{1cm} (3)$$

For small droplets initially following the flow, $Ev \ll 1$, and we have full vaporization at the flame front. For sufficiently large droplets, $Ev \gg 1$, and the droplets may slip into the flame unburned. In the present case, the $Ev$ number for droplets in the 1-5 $\mu$m range are significantly smaller than unity, so full vaporization would be expected at the flame front. However, two other effects would still be present, namely changes in the local fuel concentration, and the overall lowering of the flame temperature owing to the vaporization. The latter effect can be estimated as a sink term to the overall enthalpy. The total enthalpy per unit mass of reactant mixture required to fully vaporize the liquid droplets is given as:

$$H_{lv} = n_d \bar{V}_d h_{lv} \frac{\rho_l}{\rho_0}$$  \hspace{1cm} (4)$$

where $n_d$ is the mean number of droplets per unit volume, and $\bar{V}_d$ is the mean droplet volume and $\bar{V}_d = \int f(d) \frac{\pi d^3}{6} \, dd$ for a droplet size distribution function $f(d)$ over diameter $d$, $\rho_l$ and $\rho_0$ are the densities of the liquid and reactant mixture, and
\( h_{lv} \) is the enthalpy of vaporization of the liquid at the reactant temperature and local vapor pressure. The fraction of total enthalpy of reaction \( \Delta H \) used for vaporizing the droplet is therefore:

\[
\epsilon_{lv} = \frac{H_{lv}}{\Delta H} = \frac{h_{lv}}{\Delta H} \frac{\rho_l}{\rho_0} n_d V_d \tag{5}
\]

A corresponding maximum temperature decrease of the reactants owing to full evaporation can be calculated based on the required enthalpy, such that:

\[
\Delta T = n_d V_d \frac{\rho_l}{\rho_0} \frac{h_{lv}}{c_p} \tag{6}
\]

where \( c_p \) is the mean specific heat capacity of the reactant mixture. The effect of the lower temperature is explored by simulations presented in Section 4.3.

1.2. Effects of droplet slip motion

In practical applications, fuel droplets emerging from atomizers often move at a high velocity relatively to the surrounding air. However, in the present experiment, we consider a steady laminar flow and sufficiently small droplets, so that the droplets are dragged with a velocity comparable to the surrounding flow. These effects are normally characterized by the magnitude of the Stokes number (Stk), expressed as the ratio of inertial to viscous forces, \( \text{Stk} = \frac{\rho_l d^2 U}{18 \mu L} \), where \( U \) and \( L \) are the characteristic velocity and length, respectively; \( \mu \) is the gas dynamic viscosity. For sufficiently small Stokes numbers, slip effects should be small. In the present case, droplet diameters are of order 1–5 \( \mu m \), and the velocities are 0.9–2.2 m/s at the burner exit, and \( L \) is estimated as 0.5 mm, roughly the distance from the axial velocity turning point to the flame position, resulting in \( \text{Stk} \approx 0.001–0.2 \), less than unity. This means droplets tested in this study can approximately follow the surrounding stagnation flow, and did not produce significant slip motion. The estimation based on the Stokes
number is also supported by the prediction from previous model investigating droplet
dynamics in a counterflow flame for a droplet size of 5 µm \[19\].

1.3. Interdroplet spacing

A final thought must be given to the role of interdroplet spacing. Previous ex-
periments and models indicated that there might be an optimal spacing for each
droplet size, or conversely, a droplet size for a given concentration which maximizes
flame propagation. Mikami et al. \[25\] investigated flame propagation along a \(n\)-
decane droplet array in air to show an optimal droplet spacing that maximized the
flame propagation speed: for too sparse a concentration, the heat transferred from
a burning droplet to a neighboring unburned droplet was too slow and became the
limiting factor in flame propagation. Conversely, for high droplet concentrations,
the local flame envelope cannot provide sufficient heat for evaporation, thus lowering the reaction rate. A numerical study \[26\] summarized three flame propagation
modes that may exist for a droplet array depending on the droplet spacing. The
highest flame propagation speeds were obtained when the droplet spacing was set to
approximately four times the initial droplet diameter, which matched well with the
experimental observations in \[25\]. A recent experimental study by de Oliveira and
Mastorakos \[27\] visualized the three flame propagation modes in fuel droplets (Jet
A and a renewable alternative) and air mixtures, namely, droplet, interdroplet, and
gaseous-like flame propagation mode as the droplet concentration increases, using
OH/fuel PLIF and laser ignition. In the present study, the interdroplet spacing is
estimated as approximately 20-25 times the mean droplet diameter, depending on
the equivalence ratio. Assuming a fairly uniform distribution, at such wide distances
the group effect of droplets should be negligible.
In the following sections, the experimental method is described, and the results are discussed in the context of determining the direct effect of fuel droplets on the burning rate. Simulation results obtained from a 1D two-phase counterflow flame model at similar conditions are also presented and compared with the experimental data. The difficulty of accounting for pre-vaporization of fuel droplets and fidelity of the 1D model is discussed.

2. Experimental methods

2.1. Stagnation burner and atomization method

Figure 1 shows a schematic of the atomization method and stagnation burner system. Liquid acetone was loaded into a 50 mL volume capacity glass syringe and injected through a Delavan AL-06 atomizer. The liquid flow rate was controlled by a WPI AL-1010 high-precision syringe pump (precision 0.5%), which was carefully calibrated by a micro-scale, and delivered by a chemically resistant Tygon tube. Air flow for the atomizer was metered by a 50 slpm Alicat mass flow controller (MFC). All MFCs have a 1% full scale precision, and all air flows were pre-dried by passing through a desiccating tube filled with silica gel.

The atomizer was installed into a cylinder chamber ($φ100 \times 250$ mm, with one end aerodynamically shaped), in which the acetone mist generated was mixed with methane and the remaining air flow for a designated overall equivalence ratio $φ_o$. The flow rates of methane and air were controlled by a 20 slpm and a 50 slpm MFC respectively. The separate atomizing chamber allowed the acetone mist to mix well with the gas mixture, whilst it dampened the initial momentum of the droplets and noise generated by the atomizer. The mixture passed through a short section of 12 mm diameter tube connecting the mixing chamber to the stagnation burner.
The burner consists of a 85 mm diameter chamber, with a height of 260 mm which converges to an aerodynamically shaped nozzle of 22 mm exit diameter, shrouded by a 5 mm wide annulus of nitrogen co-flow. A water-cooled brass plate provides the impingement surface. The plate temperature was monitored by a thermocouple embedded into the plate, and controlled by tuning the flow rate of the cooling water. The body temperature of the plate was stabilized around 400 K as indicated by the thermocouple during the experiment. Another thin thermocouple was attached to the nozzle for monitoring its surface temperature. The nitrogen co-flow was controlled by a rotameter to provide the same co-flow velocity as the main flow. The distance between the burner outlet and the cooling plate was fixed at 20 mm. No liquid acetone was found to accumulate inside the mixing chamber, tube or burner even after hours of running the system. Variation of strain rates was achieved by changing the total flow rate, and thus the exit velocity of the mixture. The mean droplet size was changed by tuning the air flow through the atomizer, whilst keeping the total air flow constant.

2.2. Droplet size measurement

A Dantec phase Doppler anamometer (PDA) was aligned to measure the acetone droplet size distribution at the exit of the nozzle. The PDA system consists of an Argon-Ion laser, a 500 mm focal length transmitting probe, a 300 mm focal length collecting probe, and three photo-detectors, which were positioned at an angle of 30° off-axis from the forward scattering direction. The data rate was set to 1 kHz, and 20000 samples were obtained for each case. The thermocouple showed that the nozzle surface temperature increase was lower than 15 K for all reacting cases, so the droplet sizes should not change significantly at the burner exit. This is also evidenced by comparing the average intensity of Mie scatter which is very sensitive to the droplet
The droplet size produced by this type of air-assist atomizer depends mainly on the air blast flow rate, but was also reported to be a very weak function of the liquid flow rate [28]. The range of droplet sizes achievable with this particular setup is constrained by a compromise: a relatively long settling distance must be provided upstream of the burner exit in order to obtain a smooth velocity profile and well dispersed mist, but the final droplet size becomes smaller with distance owing to evaporation. The final droplet sizes investigated in this study are therefore below 5 µm, which are smaller than the size range studied by previous experimental and numerical work. Some acetone was present in vapor form already at the burner exit, which was estimated by subtracting the measured remaining liquid volume fraction (droplet number density times individual droplet volume) from the total fuel injected. Still, a substantial fraction of acetone vapor was produced from the droplets which were present just upstream of the flame: this means that even if vaporization takes place, the associated non-uniformity in concentration and temperature may not be dispersed in time.

The minimum value of droplet diameter that can be measured by this PDA system is about 1 µm. However in the case of the smallest droplets, a considerable fraction of droplets falls in the submicron size range, which cannot be accurately detected by the PDA system. The PDF of size distribution for the finest droplet case has a low cut off around 0.8 µm, which would bias the SMD to a larger value, as shown in Figure 2. To correct for the bias, a log normal distribution (red line) is fitted to the obtained droplet size PDF, and this fitted curve is used to determine the correct value of SMD. Figure 3 shows the corrected droplet SMD as a function of atomizing air flow rate and liquid flow rate, and the final droplet sizes (Sauter mean diameter) are determined as 4.7, 3.1, 2.2, and 1.0 µm. The droplet concentration is also monitored by the PDA system, and the result shows that changing the total
flow rate for a certain $\phi_o$ causes little difference in the droplet concentration.

2.3. **Estimation of acetone vapor fraction**

For spray experiments, one challenging but very important issue is to estimate the incoming fuel vapor fraction and quantify pre-evaporation of liquid fuel inside the burner. In the present study, the fraction of liquid acetone at the burner exit is estimated by subtracting the estimated liquid flow rate from PDA measurements $\Sigma N(d) \cdot \frac{1}{6} \pi d^3$ from the measured total liquid injection rate. Figure 4 shows such an example for an overall equivalence ratio $\phi_o = 1.0$: the measured mean droplet number density for four droplet sizes are marked by blue circles, and the theoretical value calculated from the liquid injection rate are plotted as the black dotted line. The shaded area shows the envelope of all data points. Since droplets below 0.8 $\mu$m can hardly be detected, we use the best lognormal fit (see Figure 2) to estimate the number of droplets below 0.8 $\mu$m, and added them to the measured value. The corrected droplet number density is plotted in black circles. Finally, the fraction of liquid acetone for four droplet sizes (from 1.0 to 4.7 $\mu$m) is estimated as 47%, 57%, 63% and 69% (red squares), and the corresponding acetone vapor fraction is 53%, 43%, 37% and 31%. Several liquid injection rates and total flow rates were tested; we found that the acetone vapor fraction was primarily a function of droplet size (via the air blasting flow rate), but was insensitive to equivalence ratio or total air flow rate.

PDA measurements offer very good accuracy in droplet sizing, but do not typically perform as well in measuring droplet concentrations. A precise account of the droplet concentration is only possible by optimising the PDA setup optics and laser power so as to achieve a high validation and spherical validation rate (close to 99% in all cases) as well as a high sampling rates. However, for the 1.0 and 2.2 $\mu$m SMD
cases, up to 80% and 55% of droplets smaller than the minimum measurement diameter failed to be detected by the PDA system. This was instead corrected based on the lognormal curve fitting. Nevertheless, the estimates of vapor fraction made here may have significant uncertainties. This is a common problem for spray experiments, especially for highly volatile fuels such as acetone, ethanol, and \( n \)-heptane, which are frequently used in such studies. The estimation of the vapor fraction could yield very different result depending on the assumption made. For example, de Oliveira et al. \[29\] estimated the pre-evaporated ethanol fraction from the energy balance of the flow in the burner, assuming saturated vapor pressure and thermal equilibrium has been reached due to a long residence time and adiabatic condition inside the burner. For the Sydney Burner \[30\], the vapor fraction of acetone and ethanol droplets was estimated by comparing the liquid flux measured by the PDA at the burner exit with the injection rate, same as in the present study. That paper also commented on the difficulty on measuring the liquid flux by the PDA system as significant errors and uncertainties may occur, although good repeatability of the measurement was confirmed. Earlier studies such as the Chen and Gomez \[12\] did not provide a quantitative estimation of the vapor fraction and pre-evaporation effect, but only reported an overall equivalence ratio, which must therefore be considered with extra care when comparing with model results. In general, experiments on spray combustion have larger uncertainties than gaseous flames due to pre-vaporization effects (for volatile fuels) or loss of liquid fuel (for large droplet sizes) due to deposition. Hence, it appears more practical consider the boundaries of experimental uncertainties in the model and their effects on resulting flame behavior, rather than attempting to match the exact values. In the simulation presented further on, a 50% uncertainty is considered for the vapor fraction. Instead of testing a single value, we present the results for the upper and lower bounds as well as the mean.
2.4. *PIV system*

The velocity field was measured using a 2D PIV system. Acetone droplets served both as reactant and PIV tracer. As mentioned earlier, the Stokes number for these acetone droplets is below or around unity, so they are expected to follow the flow. A double-pulsed Litron Nano-PIV laser emitting a 532 nm wavelength beam at a repetition rate of 10 Hz was used to generate a light sheet with the thickness of approximately 0.3 mm through a focal length FL = -150 mm concave cylindrical lens followed a FL = 500 mm convex lens. A 2048 × 2048 pixel CCD camera (Imager Pro X 4M, LaVision) was operated in double frame mode at 5 Hz to record the droplet image. A combination of a Nikkor 60 mm Micro lens (f/5.6D) and a Sigma ×2 teleconverter was used to focus the image. A narrow bandpass filter centered at 532 nm was installed in front of the lenses to remove flame luminosity and other background signals. The pixel resolution was 11.9 µm as calibrated by a target plate, and the final field of view was fixed at 24.4 × 24.4 mm². Example PIV images based on acetone droplets are shown in Figure. Due to out-of-plane motion and evaporation, droplet images disappeared immediately downstream of the flame front. For the finest droplet case, the droplet fully disappeared at a position about 300 µm upstream of the flame front which was indicated by the luminous zone on the second frame. Hence we used a teleconverter to achieve a sufficiently high spatial resolution to guarantee at least 2-3 rows of vectors downstream of the minimum velocity axial vector. The time interval between two laser pulses was set in the range of 80-140 µs. The images were processed using DaVis 7.2 from LaVision. Vectors were calculated using multipass cross-correlation with decreasing interrogation window size (64 × 64 for first pass, 32 × 32 for second pass, both with 25% overlap), for a 86 × 86 vector field for each image pair. In addition, the flame appearance was recorded by a Nikon D810 camera installed with a 50 mm lens (f = 2.0/D), using a shutter time of 1/60
2.5. **Determination of stretched flame speed and strain rate**

The same stagnation flame method used in a previous gas-phase study \[22\] based on \[24\] was used to calculate the stretched flame speed and strain rates. Figure 6(a) shows an example of the centerline axial velocity profile for a two-phase flame. Similarly to the flame of gaseous fuels, the axial velocity decreases towards the stagnation point, reaching a minimum velocity and then accelerating through the flame, where the acetone droplet evaporates quickly and soon disappears from the PIV image. This minimum axial velocity is defined as the reference flame speed \(S_{\text{ref}}\) at a given flow rate and equivalence ratio. Unlike the case of pure gaseous flames, the heterogeneity in equivalence ratio and temperature caused by a droplet may produce a local curvature on the flame front at a length scale of the same order of the interdroplet spacing. In the present experiment, the interdroplet spacing was below 100 \(\mu\)m, which cannot be resolved by the current imaging system. Instead, in this study, we measure an apparent flame propagation speed relative to the moving unburnt mixture. Figure 6(b) shows the radial velocity profile at the coordinate of the reference velocity. The linear gradient \(a\) of radial velocity profile is used to determine the axial strain rate \(K\) \[31\], as \(K = 2a\). A Matlab program was written to capture the reference flame speed as well as the local strain rate at the stagnation point. However, it occasionally selected an incorrect solution due to noise on the vector field. To avoid this, we monitor the results frame by frame. Finally, a total number of 200-600 data points were used to determine each flame speed slope against the strain rate.

The range of strain rates derived from this experiment is within 100-250 \(s^{-1}\), as is the original reference case, where the gas-phase flame speed data showed good
linearity. Since both single-phase and two-phase flames were only weakly stretched, the expected heat loss to the wall should be minimal [24]. Although the same method is used for measuring two-phase flame speed under a certain strain rate, we must note that, since the asymptotic theory has not been validated for two-phase opposed-jet or stagnation flames, this study does not aim at deriving the free flame propagation speed by extrapolation. Instead, the purpose is to compare the acquired velocity profiles, and eventually those of temperature, species and soot, using the reference case and simulation results under the same stretched conditions.

2.6. Operating conditions

Table 1 summarizes all test conditions. The baseline conditions in the experiments mirror those in the work of [22], in which gaseous methane and vaporized acetone are used as a fuel. We define the ratio of acetone to methane as $X_{C_3H_6O}/X_{CH_4}$ by mole, consistently with the original gas-phase study. Tests were run for 9% and 20% acetone concentration (corresponding to an energy contribution ratio of $H_{C_3H_6O}/H_{CH_4} = 18.6\%$ and 41.4\%, respectively) at overall equivalence ratios $\phi_o$ from 0.85 to 1.35. A few data sets at leaner and richer conditions were also acquired for the 9% acetone case, however under such conditions, the flame were not able to sustain for a long time. The overall equivalence ratio was calculated based on the measured total flow rates of methane/acetone/air. The achievable flammable range of equivalence ratio for the droplet-laden cases is narrower than that of the gas phase ($\phi = 0.67\text{-}1.40$), especially on the lean side. For each concentration, droplet sizes of 1.0, 2.2, 3.1 and 4.7 µm were tested. All tests were conducted at atmospheric pressure, and the ambient gas temperature varied within the range of 290 ± 2 K. The range of rich equivalence ratios and acetone/methane ratios that could be explored in this particular configuration was limited by the autoignition of droplets near the
rim of the cooling plate. For example, for \( \frac{X_{\text{C}_3\text{H}_6\text{O}}}{X_{\text{CH}_4}} = 4 \) and \( \phi_o = 3.0 \), a side flame ignited at the rim of the cooling plate where remaining acetone droplets met fresh ambient air, leading to autoignition by the high temperature burnt gases. The lower limit was bound by the ignitability of stable mixtures.

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<tr>
<th>( X_{\text{C}_3\text{H}<em>6\text{O}}/X</em>{\text{CH}_4} ) (%)</th>
<th>SMD (( \mu )m)</th>
<th>Liquid fraction (%)</th>
<th>( \phi_o (\text{--}) )</th>
<th>( Q_{\text{gas}} ) (slpm)</th>
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<td>9</td>
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<td></td>
<td>2.2</td>
<td>57</td>
<td>1.40</td>
<td></td>
</tr>
<tr>
<td></td>
<td>3.1</td>
<td>63</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>4.7</td>
<td>69</td>
<td></td>
<td></td>
</tr>
<tr>
<td>20</td>
<td>gas</td>
<td>0%</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>1.0</td>
<td>47</td>
<td>0.85</td>
<td>20-36</td>
</tr>
<tr>
<td></td>
<td>2.2</td>
<td>57</td>
<td>1.35</td>
<td></td>
</tr>
<tr>
<td></td>
<td>3.1</td>
<td>63</td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>4.7</td>
<td>69</td>
<td></td>
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</tr>
</tbody>
</table>

3. Experimental results and discussion

3.1. Flame structure

Figure [7] shows natural color flame structure images for 9% acetone concentration at equivalence ratios 0.85, 1.15 and 1.35 for the four droplet sizes. The droplet-laden flames look very different from their gas-phase counterparts (an example is also shown at the bottom of the same figure) as they show a two-layer structure: a blue flame front followed by an orange-reddish zone, whereas for the corresponding gas-phase flame, only a single flame zone was observed. There are several possible sources of this orange-reddish emission: one is radiation of soot and/or emission of \( \text{CO}^* \) or \( \text{C}_2^* \), which is consistent with the simulation predictions [17] that the flame presents both premixed-like and diffusion-like characteristics even under lean conditions. The
orange luminosity is expected to arise from the reaction around burning droplets, creating high fuel concentrations and temperatures. Interestingly, turbulent ethanol flames under overall lean conditions examined in other turbulent studies did not show an orange zone [28, 32], suggesting that the specific droplet sizes and mixing conditions may be important. The four mean droplet sizes tested are all lower than 5 µm, which are expected to fully vaporize within the reaction zone. However we still observe a significant difference in flame appearance between droplet-laden flame and their gaseous counterpart. This contradicts previous numerical predictions [16] which suggest that for very fine droplet sizes (< 10 µm), evaporation is immediate and the flame appearance and behaviour should be no different than a gaseous flame. The disparity probably comes from the assumption in numerical models that the droplet source term exists everywhere spatially, namely, the fuel vapor diffuses promptly and thus leads to a uniform mixture. In fact, the present observations suggest that under the present conditions, the mixture fraction gradients produced by the droplet vaporization may persist locally, thus producing pockets of sufficiently rich mixtures to produce either CO* or soot and increased red-shifted luminosity for C2* swan bands. For the very rich case, \( \phi_o = 1.35 \), the flame has a green tinge, indicating soot may starts to form also in the first stage region. Yet, no soot was found to accumulate onto the plate surface, consistent with a stagnation configuration observed in [15]. Another possible origin of the orange hue may be the oxidation of hydrogen, which was produced during the pyrolysis of hydrocarbon fuels. According to the work by Schefer et al. [33], highly vibrationally excited H2O formed during hydrogen oxidation has a reddish emission from 600 to about 900 nm, which may also contribute to the colour shown in Figure 7.

For smaller droplet sizes and correspondingly reduced interdroplet spacing, the luminosity of the orange zone increases, whilst for the larger droplet size of 4.7 µm,
the orange zone starts to disappear. This is not surprising, as the observed colour is an integration of the emission from all the droplets/fuel rich pockets across the flame. Hence, the intensity of the reddish emission is directly related to the droplet number density. For the finest droplet case, the droplet number density is 1-2 orders of magnitude higher than that in the largest droplet case; consequently, the reddish colour in the finer and richer case appears to be more prominent. Further, reactions inside the rich pockets generated by a large droplet may not be complete, owing to slow mass diffusion and a low local temperature caused by vaporization; both factors would be consistent with lower emission of radiation or chemiluminescence than in the case of finer droplet mists.

As the overall equivalence ratio increases from 0.85, the stagnation point gradually moves upstream towards the nozzle and reaches a closest position to the nozzle at $\phi_o = 1.15$. Further increases to $\phi_o$ lead the flame downstream towards the cooling plate until either the flame extinguishes or a side diffusion flame is formed at $\phi_o$ around 1.42, depending on the shielding nitrogen flow rate. The flame produces acoustic noise under higher power rich cases and high flow rates, as also reported in [15] for a similar stagnation setup. The fluctuations may be caused by the flapping edge of the flame, which can be seen in some of the pictures in Figure. However, the noise has little effect on the central region of the flame front, where measurements were made.

3.2. Laminar flame speed as a function of strain rates

For gaseous flames, a linear relation between burning velocity and strain rate has been derived theoretically by Matalon and Matkowsky’s early work in the 1980s
and validated experimentally by Wu and Law \cite{36}:

\[ S_{\text{ref}} = S_0 - \mathcal{L}K \]  

where \( S_0 \) is the free flame speed at zero strain rate, \( \mathcal{L} \) is the Markstein length, and \( K \) is the strain rate. Later, Tien and Matalon \cite{37} pointed out that for the opposed-jet/stagnation flame method, a non-linear relation between the measured reference velocity and strain rate could be expected. Therefore a non-linear extrapolation was proposed for this method to reduce the biases in deriving the free flame speeds. In our previous acetone/methane experiment \cite{22}, the non-linear effect was found to be trivial as the Karlovitz number was very low (smaller than 0.1) \cite{23}, so the measured data points could be fitted by a linear relation quite well. However, all previous analyses did not consider fuel in liquid form, hence the relation between flame speed and strain rate still remains unknown when droplets are present. The experimental results in this study for the first time reveal this relation for two-phase flames.

Following the method introduced in Section 2.5, reference axial burning velocities \( S_{\text{ref}} \) are extracted from each PIV image pair, along with the corresponding strain rate, as shown in Figure 8 for (a) \( \alpha = 9\% \), and (b) 20\%. The full data sets are presented in the supplementary materials (Figure S1 and the csv file). The values are shown as scatter plots of \( S_{\text{ref}} \) as a function of strain rates \( K \), for a range of equivalence ratios and droplet sizes. A best fit slope is shown as a black line on the subplots. The mean flame speeds for the fully vaporized case are shown as red dashed lines, obtained in \cite{22}. The random fluctuation in the measured flame speeds (\( \sim 2 \text{ cm/s} \)) is slightly higher for the droplet-laden cases than that of the fully vaporized flame (1-2 cm/s). The latter falls in the typical fluctuation level of gas-phase flame speed measured by PIV, as reported, for example, in \cite{38}. This may arise from the occasionally very
large droplets (> 20 µm from the PDA result) penetrating through the reaction zone and disturbing the flame front, and the residual acoustic noise from the air blast atomizer.

Considering the behavior of fully vaporized flames with equivalence ratio first (dashed red lines), we observe that the peak reference flame speeds are obtained at equivalence ratios around 1.05. The slope of the dependence of laminar flame speed with strain varies from positive in the lean case to flat as the flame becomes richer.

For flames with droplets, at an equivalence ratio around 1.15, both the fully vaporized and the droplet-laden flames show very similar reference flame speeds and dependence on strain. On the lean and rich side, however, the behavior deviates in opposite directions: the absolute value of the flame speed with droplets is lower than the fully vaporized mixture in the lean case, yet higher in the rich case. Further, whilst the slope of the reference velocity with strain on the lean side is positive and approximately the same the vaporized case, the presence of fine droplets (1.0 and 2.2 µm) on the rich side makes the slope with strain negative rather than flat as in the vaporized case.

The Markstein length, \( L = -\frac{dS_{ref}}{dK} \), is used to describe the relationship between the flame speed and local strain rate for gaseous flames. This parameter plays an important role in turbulent flamelet simulations. However, since the two-phase flamelet theory is yet to be thoroughly developed, here we use the word “gradient” instead of Markstein length to represent this relation. The slope gradients are extracted from Figure 8 and plotted against the overall equivalence ratio for 1.0 and 4.7 µm droplet in Figure 9. The gradient of the flame speed slope strongly depends on the droplet size \( d \): for \( d = 4.7 \) µm, the gradient of flame speed slope behaves similarly to the gas phase case: they gradually decrease and converge to zero. However, for \( d = 1.0 \) µm, the gradient crosses zero at \( \phi_o = 1.18 \) and becomes to a negative
value. These observations are consistent in both 9% and 20% acetone cases. This suggests that even very fine fuel droplets significantly change the behavior of the flame to strain rates, as is also consistent with the observations for the flame appearance in Figure [7]. Previous numerical simulations [16] stated that if the droplets are sufficiently small and fully vaporized before the flame front, the droplet-laden flame behaved no differently against their gas-phase counterpart. However, as shown by the 1 µm case tested in this study, even though approximately 53% acetone remains in vapor form at the burner exit and all remaining droplets disappear before reaching the flame front, the flame speeds and their response to strain rates appear to be very different than those measured from premixed gaseous cases. This again suggests that the mixture fraction and temperature gradient in space caused by local evaporation may have a significant influence on the flame propagation in spray clouds, which has not been captured by the flame simulations with point-source droplets. According to our experimental results, for a droplet-laden mixture under a slow mixing condition, for instance in a quiescent mist or in a laminar two-phase flow, even if all droplets were fully vaporized before reaching the flame front, the flame behavior could still be very different from its well-premixed gaseous counterpart. This feature cannot be predicted by 0D or 1D models which do not consider mixing, but possibly could be eventually captured in a high-resolution Direct Numerical Simulation (DNS).

3.3. Negative slope of flame speed

For gaseous flames, it is not unusual to observe a negative flame speed slope (or a positive Markstein length) in a counterflow or stagnation configuration. As thoroughly analyzed in [36–39], the influence of stretch on the flame speed is directly associated with preferential diffusion: increasing the flow rate in a counterflow configuration to achieve higher strain rate means providing more fresh gas to the
flame, yet it also cools down the flame due to heat diffusion to the fresh gas. The heat and mass transfer are competing in this mechanism. Hence the effective Lewis number $Le$, which is defined as the ratio of thermal diffusivity to mass diffusivity, determines the behavior of flame speed with respect to stretch. The negative flame speed slope is usually observed in a counterflow flame for a mixture with $Le > 1$, for example, lean combustion of heavy hydrocarbons [36]. There has been a vast number of studies on the Markstein number [37, 40, 41] and ways to calculate the effective Lewis number of the gas mixture [42]. For two-phase flames, adding water droplets to a propane/air [43] or a methane/air [44] stagnation flame appears to also have turned a positive flame speed slope against strain rate to negative. Therefore, the inhibiting effect of strain rates on the flame speed at rich conditions observed in the present study is probably also related to the cooling effects caused by evaporation of acetone droplets, which act as a heat sink during the early stage of reaction. It can also explain why the negative slope was found in the finer droplet cases, as the evaporation is immediate, whilst larger droplets experienced longer evaporation time in the reaction zone. Furthermore, larger droplets were only sparsely distributed in the flow as shown in Figure 5(a), local evaporation may be insufficient to affect the global flame speed. Hence, in the 4.7 $\mu$m droplet case, the flame behaved similarly as a gaseous flame (at $\phi_e$ as introduced further on) to increasing strain rate.

3.4. Enthalpy difference

One of the important effects associated with the presence of droplets is the required energy for their evaporation. The energy load is proportional to the total liquid fraction, as expressed in Eqs. 5 and 6. The difference in total enthalpy, or the temperature drop due to vaporization could be one possible reason that causes the disparity between the two-phase flame and its gaseous counterpart. Considering the
cases with the lowest and highest liquid loads, respectively: 9% acetone at equivalence ratios 0.85, and 20% acetone at equivalence ratio 1.35. The enthalpy change of vaporization for acetone is 31.3 kJ/mol. The necessary enthalpies of absorption for the different mixtures are estimated between 8.2 and 23.5 J per litre of gaseous mixture. Assuming that all droplets have fully vaporized, the corresponding overall temperature drop in the gas mixture due to acetone evaporation is estimated between 7 and 20 K, based on air properties (an estimate based on air-fuel properties would yield slightly lower values on account of the higher specific heat for vaporized acetone). Such temperature decreases are consistent with a lower flame speed. The effects of enthalpy difference are further investigated and discussed in Section 4.3 on numerical simulations.

3.5. Effect of droplets on stretched flame speed curves

Figure 10 summarises the mean reference flame speeds as a function of overall equivalence ratio $\phi_o$ for a range of strain rates for both 9% and 20% acetone cases. These are extracted from the same data as Figure 8, averaged, and replotted in a different coordinate. Only the two extreme cases in droplet size (1.0 and 4.7 µm) are plotted, whilst the results for 2.2 and 3.1 µm cases sit between those two cases. The curves for $K = 0$ s$^{-1}$, and the rich side of those for $K = 100$ s$^{-1}$ were obtained by linear extrapolation. Again, we emphasize that the linear extrapolation method has not been validated for two-phase flows, hence we do not claim that the reference flame speed extrapolated towards zero strain rate is the actual free flame propagation speed in the droplet-laden mixture.

Compared with the reference gas phase case, the curves for droplet-laden flames shift towards the richer range: lower flame speeds on the lean side, and higher on the rich side. The reference case shows a maximum flame speed at $\phi_o = 1.05$, whereas the
flame speed for the droplet-laden flame peaks at about $\phi_o = 1.15$ (except for the 1.0 $\mu$m case at $K = 0 \text{ s}^{-1}$), which coincides with the flame location nearest the nozzle (Figure 7). A physical explanation for the phenomenon is associated with a two-stage process: the energy provided by oxidation of pre-vaporized fuel at the flame front is used to vaporize the remaining liquid. This results in a reaction at a lower effective equivalence ratio $\phi_e < \phi_o$ at the first stage. The eventually vaporized acetone reacts with the remaining oxygen at a richer condition, $\phi_e > \phi_o$, downstream as the second stage. Even for the finest droplet case where all droplets should have been fully vaporized before the flame front, the local evaporation may introduce fuel rich pockets in the flow field. The fuel vapor inside these pockets may not be consumed instantaneously at the flame front due to a lack of oxygen. This also leads to leaner combustion at the flame front. In the present experiment, the effective equivalence ratio $\phi_e$ may be closest to unity at $\phi_o = 1.15$. This would explain why the flat flame front (first-stage reaction) still remains light blue for an overall rich condition $\phi_o = 1.15$, as shown in Figure 7. As a counteracting effect, according to simulations by [16], intermediate species such as hydrogen and acetylene produced during the rich oxidation of the liquid fuel (in the case of $n$-heptane droplets) may diffuse towards the oxidation zone and enhance the reaction rate. A similar mechanism may exist in the present case.

Although the right-shift of flame speed curves can be explained by introducing the concept of an effective equivalence ratio, we notice that a contradictory observation to the present observations was reported by [9], where the flame speed curve for ethanol droplets was shifted towards the lean side. However, in that study the flame speed measurements were conducted at 0.2 MPa and under microgravity conditions, which may change the underlining physics and lead to different results.
3.6. Effect of droplet size

For the 1.0 \( \mu \text{m} \) droplet case at \( K = 0 \) and 100 s\(^{-1}\), the flame speed on the rich side rises to a surprisingly high value (see the red circles on the left column in Figure 10), for example, about 50 cm/s for 9% acetone at \( K = 0 \) s\(^{-1}\). This is of course caused by the extrapolated negative slope as shown in Figure 8. Whether such a high flame speed at very rich conditions has any physical meaning is not clear yet, but this monotonic-increase shape of the flame speed curve coincides with the numerical prediction on the free flame speed in quiescent \( n \)-heptane and \( n \)-decane sprays by Neophytou and Mastorakos [16]. Further theoretical framework needs to be completed before drawing any conclusions regarding the flame propagation speed at zero strain rate. Another interesting observation from Figure 10 is that the flame speed curves are sensitive to droplet size only at low and medium strain rates. At high strain rates (200 s\(^{-1}\)), the flame speed curves for all four droplet sizes become very close.

3.7. Effect of liquid loading

Flame speed curves for 4.7 \( \mu \text{m} \) droplets for 9% and 20% acetone at 200 s\(^{-1}\) are extracted and compared with gaseous cases, as shown in Figure 11. First we consider the change between a methane-air flame and a methane-acetone-air flame, with no droplets present. A minor shift towards the lean side and a higher peak can be observed from 0% to 9% acetone; however, little difference can be observed between from 9% to 20% case, except on the rich side. The presence of droplets replacing the vapor leads to a clear shift in the overall curve towards the rich side compared to the vapor-only curve. This is consistent with the idea of an effectively lower equivalence ratio corresponding to the vaporized fraction of the fuel participating in the reaction. However, the curve is shifted downwards to a lower value, which cannot be explained
solely by the effective equivalence ratio theory. One possible reason may be the enthalpy difference mentioned earlier: as the droplet loading further increases, the heat sink effect of liquid acetone droplets as they vaporize starts to dominate. This was also pointed out in [9]: for an overall equivalence ratio $\phi_o = 0.8$, the flame speed peaked at a liquid equivalence ratio of around 0.15 for ethanol (i.e. liquid/gaseous ethanol molar ratio $X_l/X_g = 23\%$).

In the same study, further increasing the liquid ethanol loading lowered the flame propagation speed. Smaller interdroplet distances also reduced the flame speed when the liquid loading was high, as concluded in [25]. However, the two-phase counterflow flame simulation presented in the following section does not consider the temperature gradient between individual droplets. Instead, the simulation calculated the overall temperature drop in the gaseous mixtures, and hence predicts a limit of the heat sink effect. A more accurate prediction would require resolving more finely the droplet evaporation and the local scalar fields, in order to reflect the rate of burning in a droplet cloud.

4. Numerical simulations of two-phase counterflow flames

In this section, we introduce the numerical method for a two-phase stagnation flame, and the simulation results. The simulations use a specially developed two-phase counterflow flame module in COSILAB [45], which extends the capabilities for the free-flame calculations described in [16] to stagnating flames. Egolfopoulos and co-workers [24] suggested that the symmetric counterflow and stagnation models for the 1D flame calculations produced very similar result at low strain rates (up to $150 \text{ s}^{-1}$ for lean, and $300 \text{ s}^{-1}$ for stoichiometry and rich conditions for methane/air flame). However, at high strain rates, the stagnation model may underestimate the flame speed, because the flame front is pushed very close to the wall, such that the
adiabatic temperature cannot be achieved due to heat loss. The slopes shown in Figure 8 do not indicate a sudden drop in the strained flame speed at high strain rates as observed in [24]. It is therefore reasonable to claim that near to adiabatic conditions was achieved in the experiment, and that the results can be reproduced by a counterflow flame configuration with symmetric boundary conditions on both sides for the tested range of strain rate.

4.1. Numerical method

The methodology and governing equations for the two-phase counterflow flame model in COSILAB were documented in a previous paper from the code developers [21], which followed the same Eulerian-Lagrangian method and similarity formulation as used in [18–20]. The governing equations and boundary conditions are described in Appendix A. The two-phase flame model uses the similarity formulation for the 1D gas-phase counterflow flame model, and adds a droplet evaporation model, which links the two phases via the source terms in mass, species, momentum and energy due to droplet evaporation and motion. The gaseous flame solver provides a temperature profile to the liquid solver to calculate the evaporation process, the latter then returns the updated profiles of fuel vapor and temperature to the former for a new round of gaseous flame calculation. The two solvers are running alternatively until final convergence has been reached. Due to the non-linearity of the problem, and the fact that a two-phase mixture may only produce a stable solution for a narrower range of equivalence ratios and strain rates comparing with gas mixtures, convergence in a two-phase counterflow flame model is much more difficult to achieve than in the single-phase model. The calculation starts with an input initial guess on a set of profiles for all the parameters such as velocity, temperature and major species, which must be sufficiently close to the final solution in order to converge. The equivalence
ratio, strain rate, or the droplet size was varied in small increments from one case to the next, and the last computed solution was used as the next initial guess.

4.2. Mechanism and gas-phase counterflow flame simulation

One major advantage of 1D two-phase counterflow flame modeling is that it incorporates chemistry into the simulation, which gives a more realistic interpretation on the phenomenon and can be directly compared with experimental results for a specific fuel. The acetone and methane reaction mechanism used here is consistent with the one proposed in our previous study [22]. Seven reversible reactions involving acetone and intermediates were added to GRI-Mech 3.0 [46], see Table 2.

<table>
<thead>
<tr>
<th>Reactions</th>
<th>$A$</th>
<th>$n$</th>
<th>$E$</th>
<th>Ref.</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\text{CH}_3\text{COCH}_3 \leftrightarrow \text{CH}_3\text{CO} + \text{CH}_3$</td>
<td>7.02E+21</td>
<td>-1.57</td>
<td>84,684</td>
<td>47</td>
</tr>
<tr>
<td>$\text{CH}_3\text{COCH}_3 + \text{H} \leftrightarrow \text{CH}_3\text{COCH}_2 + \text{H}_2$</td>
<td>9.80E+05</td>
<td>2.43</td>
<td>5162</td>
<td>47</td>
</tr>
<tr>
<td>$\text{CH}_3\text{COCH}_3 + \text{O} \leftrightarrow \text{CH}_3\text{COCH}_2 + \text{OH}$</td>
<td>1.00E+13</td>
<td>0.00</td>
<td>5961</td>
<td>48</td>
</tr>
<tr>
<td>$\text{CH}_3\text{COCH}_3 + \text{OH} \leftrightarrow \text{CH}_3\text{COCH}_2 + \text{H}_2\text{O}$</td>
<td>1.25E+05</td>
<td>2.48</td>
<td>445</td>
<td>47</td>
</tr>
<tr>
<td>$\text{CH}_3\text{COCH}_3 + \text{CH}_3 \leftrightarrow \text{CH}_3\text{COCH}_2 + \text{CH}_4$</td>
<td>9.50E+03</td>
<td>2.50</td>
<td>8400</td>
<td>49</td>
</tr>
<tr>
<td>$\text{CH}_3\text{COCH}_2 \leftrightarrow \text{CH}_2\text{CO} + \text{CH}_3$</td>
<td>1.00E+13</td>
<td>0.00</td>
<td>28,000</td>
<td>49</td>
</tr>
<tr>
<td>$\text{CH}_3\text{CO} + \text{M} \leftrightarrow \text{CH}_3 + \text{CO} + \text{M}$</td>
<td>5.36E+27</td>
<td>-3.40</td>
<td>18,900</td>
<td>50</td>
</tr>
</tbody>
</table>

The free flame propagation simulation result using this mechanism matched very well with the free acetone flame speed measurements obtained by extrapolation, as reported in [22]. Before starting the two-phase simulation, we conduct the simulation for a gas-phase counterflow flame using this mechanism for 0%, 9%, 20%, and pure acetone cases in COSILAB, and compare with the experimental data, as shown in Figure 12. Detailed description on the gas-phase counterflow flame model can be found in [51]. The markers show the experimental data extracted from our previous work, and the lines correspond to the numerical result. Solid lines are cal-
culations based on the adapted GRI-Mech 3.0 shown in Table 2. For comparison, in Figure 12(b)-(d), the simulation results based on another acetone mechanism introduced by Pichon et al. [47] are also plotted as dashed lines. The strain rate for simulation ranges from 80-250 \( s^{-1} \), to overlap with the conditions over which the experiment was conducted. Within this range, a linear relationship between flame speed and strain rate is supported by the simulation result. The calculations offer reasonable predictions for the strained flames, except in the lean region (blue), where the calculations based on the adapted GRI mechanism overestimate the flame speed of the blends. Notice that this is a similar result as obtained by a free flame simulation [22] in the same range for the gaseous flames. The gas-phase simulation results provide a clean baseline for the following two-phase counterflow flame simulations. However, there are some clear discrepancies associated with the slope of the curve in the lean case (blue), as well as for the lean case in pure acetone, which is systematically lower than the calculated values.

4.3. Two-phase counterflow simulation

In the two-phase counterflow simulation, we reproduced 1.0 and 4.7 \( \mu m \) droplet cases for three different equivalence ratios: 0.85, 1.05, and 1.3, respectively. Figure 13 shows the axial profiles for 20% acetone, 4.7 \( \mu m \) case near stoichiometry. Contrary to observations, the simulation predicts that all droplets fully evaporate far ahead the flame front, as shown in Figure 13(a). The molar fraction of acetone vapor correspondingly reached a plateau before the reaction started, see Figure 13(b). Therefore, the simulation results should be very similar to those given by a gas-phase counterflow flame simulations at the same condition, except that the simulations predict a 16 K temperature drop in the gas mixture due the liquid acetone evaporation, as indicated in Figure 13(c). Figure 13(d) shows the axial velocity profiles.
The numerical prediction on droplet evaporation contrasts with the experimental observation in Figure 5: acetone droplets did not fully disappear until they reached the flame front. One possible reason to explain this disparity is that the above simulations start with acetone in a purely liquid phase, and predict shorter vaporization time than in reality, where substantial vapor is expected to be available at the nozzle (Section 2.3). To investigate the possibility, the acetone vapor concentration has been estimated for each case in Section 2.3 and simulations were reconducted with acetone in both vapor and liquid form at the boundary from 0 to 50% for the 4.7 µm droplet case, to account for uncertainties in the possible fractions. The profiles of acetone vapor molar fraction are presented in Figure 14. Again, all liquid acetone is predicted by the code to vaporized entirely at about 5 mm above the nozzle, far ahead the flame front (13 mm above the nozzle). The initial vapor fraction due to pre-evaporation seems to play a trivial role on droplet evaporation. Clearly, the model is at odds with the experimental data, and we expect that the main reason may be the discrepancy in the mass and heat transfer number, $B_T$ and $B_M$, at the boundary, which does not consider the local high vapor concentration as well as the low temperature in the gas film due to pre-vaporization. This clearly would overestimate the rate of vaporization. Another reason could be that the model used here is a similarity model of the boundary layer type which assumes that transverse to the stagnation point streamline the thermal structure of the flame (in terms of density, temperature, mass fractions) does not vary. As a consequence, the gas-phase model is valid only in the vicinity of the stagnation point streamline. The model may not be able to capture all the features for a 2D field.

Unlike the current 1D model, in reality droplets are not point sources, but sources of the order of up to 5 µm in diameter, whereas the grid sizes in the calculations are of the order of a few tens of µm to resolve the flame gradients. This means
that concentration gradients arise over distances of the same order of magnitude as the diffusive/evaporative layer of the droplets. This breaks the assumptions made in the derivation of the Spalding number, and means that a more resolved model than a point source may be required to capture the nuances of droplet-droplet and droplet-mixture interactions. Further higher resolution direct numerical simulations may be required to resolve droplets and their interactions near the flame.

In spite of the challenges in reproducing the exact evaporation process for the current model, it is still possible to make comparisons between the numerical results and experimental data, which are shown in Figure 15. For the simulation results, the minimum axial velocity was extracted as the reference flame speed, whilst the axial velocity gradient ahead of the velocity turning point was used as the local strain rate. Figures 15(a) and (c) show that, as expected, the results of the gas phase simulations become identical to the droplet simulations, once their respective temperature and enthalpy drops are accounted for. Nevertheless, neither of these calculations entirely captures the change in reference speed in the observations: the simulated reference speeds only slightly overestimate the expected values in the lean case, even though the observed slope gradient with strain is higher than the simulations. The stoichiometric cases capture the slope well, but values are underpredicted. In the rich cases, slopes are reasonably predicted for the droplet size 4.7 µm, but the values are severely underpredicted. Clearly further work is required to resolve some of these new observations, and the authors are planning collaborations on the matter.

5. Conclusions

We considered experiments of laminar stagnation mixtures of methane/air with added acetone in either gas or droplet phases, and compared the resultant reference flame speeds of the otherwise identical mixtures as a function of strain rate,
global equivalence ratio and acetone concentration. The flame propagation speed under various strain rates from 100-250 s$^{-1}$ was measured using PIV, using the small droplets as tracers. Cases with four mean droplet sizes (SMD 1.0, 2.2, 3.1, 4.7 $\mu$m), two acetone concentrations (9% and 20% by mole), and equivalence ratios between 0.80 to 1.35 were tested. The reference flame speeds obtained for the droplet-laden cases were compared with a reference case where all acetone was vaporized. Simulations using well-established counterflow single- and two-phase flame models were conducted and the results were compared with the experimental data. The main findings are summarized below:

The droplet-containing flames show a two-layer structure even for droplet size as small as 1.0 $\mu$m. A thin blue flame front followed by a thick reddish zone was observed. This suggests that the mixture reacted in a two-stage mode: at the flame front, methane and gas-phase acetone was oxidized at locally lean conditions ($\phi_e < \phi_o$), whilst in the post flame zone the acetone vapor in locally fuel-rich pockets can react with the remaining oxygen ($\phi_e > \phi_o$). This leads to a flame with both premixed-like and diffusion-like characteristics as suggested by previous computational work.

Under lean conditions, droplet addition significantly decreases the reference flame speed relative to fully vaporized conditions, indicating a lower reaction rate, even for very small droplet sizes, and at all strain rates. The sensitivity to strain rate is not significantly affected. This appears to be accounted for largely by the enthalpy of vaporization of the droplets, according to 1D strained flame model results with droplets and gas phase model results at lower initial temperatures corresponding to the full vaporization of the droplets.

Under rich conditions, the stretched flame speeds of droplet-laden flames increase significantly, relative to the gas phase counterparts, at all strain rates. Their sensitivity to strain is low or (in the case of very small droplets) negative, in contrast
with the fully vaporized counterpart. This is surprising, and cannot be predicted by current 1D strained flame model, with or without point-source droplets: the former predicts a decrease in the flame speed, owing to the lowered temperature. It may be possible that the presence of droplets in an overall leaner mixture creates locally rich regions, which in turn leads to higher than average reaction rates and temperatures. This would explain both the two-zone appearance of the flame, and the increase in stretch flame speeds. Further experimental and modeling evidence is clearly required to completely understand the problem.

A method has been generated to create and examine the behavior of steady, droplet-laden laminar strained flames based on two fuels (methane and acetone). The method can be adapted to a number of fuels and mixtures relevant to spray combustion studies and submodels, including the deployment of further optical diagnostic techniques to probe the reacting region.

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Appendix A. Governing equations

The equations for gas and liquid phase are solved as a one-dimensional axisymmetric system about the axial flow direction. A volumetric void fraction is defined as

\[ \epsilon = \frac{\rho_f}{\rho_g} \]  

(A.1)

where \( \rho_f \) and \( \rho_g \) are the mass of gas per unit total volume, and \( \rho_g \) is the gas density at the local pressure, temperature and composition. The gas phase equations are handled in Eulerian and Lagrangian frameworks, respectively, and are described in summary form below.

Gas phase. We define the two-dimensional operator :

\[ L(\varphi) = \frac{\partial \rho_f \varphi}{\partial t} + \frac{1}{x_j} \frac{\partial \rho_f u_g \varphi}{\partial x} + \frac{\partial \rho_f v_g \varphi}{\partial y} \]  

(A.2)

where \( j = 1 \) for the current radially symmetric problem, \( y \) is the longitudinal (co-flow) and \( x \) the transverse coordinate, and \( v_g \) and \( u_g \) are the corresponding gas phase velocities. So the governing equations for mass, species, momentum and energy read:

\[ L(1) = S_m \]  

(A.3)

\[ L(Y_{g,i}) = -\frac{\partial (\rho_f V_{g,i} Y_{g,i})}{\partial y} + \epsilon w_i + S_{m,i} \]  

(A.4)

\[ L(v_g) = -\frac{\epsilon p}{\partial y} + \frac{\partial}{\partial y} \left( \epsilon \mu_g \frac{\partial v_g}{\partial y} \right) + S_v \]  

(A.5)

\[ L(h_g) = -\frac{\epsilon q_g}{\partial y} - \epsilon \sum_i h_i w_i + S_e \]  

(A.6)

The variable \( Y_{g,i} \) is the gas phase mass fraction and enthalpy, and \( V_{g,i} \) the corre-
sponding diffusive velocity, $p$ the local pressure, $h_g$ the gas enthalpy, $w_i$ the reaction rate of species $i$, and $\mu_g$ the gas viscosity. The term $q_g$ is the $y$ component of the gas phase heat flux, calculated as:

$$q_g = -\lambda_g \frac{\partial T_g}{\partial y} + \sum_{i=1}^{I} j_{g,i} h_{g,i}$$  \hspace{1cm} (A.7)

where $T_g$ is the gas phase temperature, $\lambda_g$ the gas phase conductivity, and $j_{g,i} = \rho_f V_{g,i} Y_{g,i}$ the diffusive flux of species $i$.

The remaining source terms denoted by $S$ result from the liquid phase, and are defined as follows:

$$S_m \equiv n \dot{m}$$  \hspace{1cm} (A.8)

where $n \dot{m}$ is the total mass evaporation rate of droplets from liquid to mass phase, and the current formulation assumes a stream of monodisperse droplets. We note that for these small droplets, oscillatory motion of droplets overshooting the flame and returning is not important.

$$S_v \equiv n \left( \dot{m} v_d - m_d \frac{dv_d}{dt} \right)$$  \hspace{1cm} (A.9)

where $m_d$, $\dot{m}$ and $v_d$ are the droplet mass, rate of evaporation and velocity of the droplets.

$$S_e = -n[\dot{q} + \dot{m}(C_{Pd}(T - T_s) + L_v)]$$  \hspace{1cm} (A.10)

where $\dot{q}$ is the heat transfer rate required for the evaporation of the droplet, $C_{Pd}$ is the heat capacity of the liquid, $T_s$ is the surface temperature of the droplet, and $L_v$ is the latent heat of evaporation.

**Liquid phase.** The droplet mass evaporation rate and corresponding energy rate
required are given as:

\[ \dot{m} = \pi \rho_f D_f d Sh \ln(1 + B_M) \] (A.11)

where \( D_f \) is the local diffusion coefficient, \( d \) the local droplet diameter, \( Sh \) the local Sherwood number and \( B_M \) the local Spalding evaporation number,

\[ B_M = \frac{Y_{f,s} - Y_{f,\infty}}{1 - Y_{f,s}} \] (A.12)

The rate of energy transfer to the droplet for evaporation is:

\[ \dot{q} = \dot{m} \left[ \frac{C_P d(T - T_s)}{B_T} - L_v \right] \] (A.13)

where the factor \( B_T \) accounts for the differential diffusion of mass and energy around the droplet. Details of all the property calculations regarding droplet evaporation are referred to the COSILAB manual details [45] or the original paper on this evaporation model [52].

The droplet trajectory of an individual droplet is expressed by \((x_d(t), y_d(t))\). For convenience we define a slip velocity vector.

\[ \mathbf{v}_s \equiv (u - u_d)i + (v - v_d)j \] (A.14)

Based on Newton’s Second Law, the droplet equations of motion can be written as

\[ m_d \frac{\partial^2 x_d}{\partial t^2} = \frac{1}{2} \rho |\mathbf{v}_s| \pi r^2 [-C_L(v - v_d) + C_D(u - u_d)] \] (A.15)

\[ m_d \frac{\partial^2 y_d}{\partial t^2} = \frac{1}{2} \rho |\mathbf{v}_s| \pi r^2 [C_L(u - u_d) + C_D(v - v_d)] + m_d g \] (A.16)

where \( r \) is the droplet radius, \( C_L \) and \( C_D \) are the lift and drag coefficient, respec-
tively. Note that here gravity is considered for the axial direction, appear as $m_d g$ in Eq. (A.16). Again, since very fine droplets are investigated in this study, the effects of slip velocity and lift/drag force should be trivial, as will be shown later on. For a steady droplet velocity field $V_d$, the flux of droplet number density is conserved,

$$\nabla \cdot (n V_d) = 0 \quad (A.17)$$

Integrating the above equation and based on a similarity analysis, the droplet number density is expressed as

$$n = \frac{n_0 x_d v_d}{x_d v_d} \quad (A.18)$$

where $n_0$, $x_d$, and $v_d$ are the initial values at the cold boundary.

Boundary conditions The governing equations for the gas and liquid phase form a set of $3 + K$ gas phase equations, for the variables $(u_g, v_g, Y_{g,i}, \frac{dp}{dy}, h_g, \rho_f, \epsilon)$. The closure is provided by the $(3 + K)$ matching conditions for mass, species, momentum and energy conservation between phases, and the additional 3 equations for the droplet motion and conservation equations correspondingly for $(x_d, y_d, n)$.

Boundary conditions in the present quasi-steady flow case are by potential flow conditions at the outer boundary for $u_g, v_g$, with identical velocities for the droplet phases, assumed to be distributed uniformly in the non-reacting boundary, measured conditions for the reactant gas species $Y_{g,i}$ and temperatures, and zero gas phase concentrations for the fuel species at the reactant boundary. The longitudinal pressure gradient is approximated as usual in the boundary layer approximation by the imposed pressure gradient at the potential flow. Symmetry conditions for all gas-phase variables are assumed at the downstream boundaries.
Figure 1: Schematic view of the atomization and burner system
Figure 2: Sample droplet diameter PDF acquired by the PDA for two diameter settings. The bins are cut off below 0.8 µm, as limited by the resolution of the instrument, which biases the Sauter Mean Diameter (SMD) to a larger value. The best lognormal distribution (red line) is fitted to the obtained data to calculate the SMD.
Figure 3: Sauter Mean Diameter (SMD) of acetone droplets measured by the PDA system at the burner exit, as a function of air flow rate through the atomizer. Three different liquid flow rates $Q_l = 0.7, 2.0$ and $3.4$ ml/min were tested. The SMD is mainly dependent on the air blast flow rate, but changes negligibly with liquid injection rate $Q_l$. 

![Graph showing SMD vs. Air blast flow rate]
Figure 4: Estimated liquid acetone fraction at the burner exit obtained by subtracting the total liquid volume obtained from PDA measurements over the bins ($\sum_{i=1}^{N} n_i \cdot \frac{1}{6} \pi d_i^3$) from the total liquid injection rate. The blue circles represent the mean droplet number density measured by the PDA system, the shaded area marks the error range envelope of data points. The corrected droplet number density is plotted in black circles, after lognormal fitting (see Figure 2). The black dotted line shows the measured total liquid concentration as calculated from the injection. The final fraction of liquid acetone for four droplet sizes is obtained by the subtraction of the corrected liquid concentration from the measured total liquid injected. The final liquid fractions are shown as red squares for 1.0 to 4.7 $\mu$m.
Figure 5: Example PIV images (second frame, zoom-in view) for droplet sizes at overall equivalence ratio $\phi_o = 1.0$. The total gaseous flow rate was 30 slpm. The flame front position is marked by the flame luminosity due to the long exposure time of the second frame. The contrast between particle image and flame luminosity was adjusted for better display.
Figure 6: (a) Example of velocity field measured by PIV for the two-phase hybrid gas-liquid flame. (b) Single-shot axial and (c) radial velocity profiles extracted from (a). The local burning velocity is taken as the minimum axial velocity, and the strain rate is taken as twice the radial velocity gradient $K = 2a$. 

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Figure 7: Flame appearance at $\phi_o = 0.85, 1.15$ and 1.35 for 9% acetone. The total gaseous flow rate was 30 slpm. Pictures of gas-phase acetone/methane flame at similar condition are also presented in this figure. The images for lean gas cases produced in [22] are not available.
Figure 8: Reference flame speeds for the case (a) $\alpha = 9\%$ and (b) 20\%, for a range of global equivalence ratios (columns) and mean droplet sizes (rows). Black lines indicate best fit lines, red dotted lines the corresponding fully vaporized case.
Figure 9: Gradient of reference flame speed slopes (unit: mm) extracted from Figure 8 as a function of $\phi_o$ and droplet size. Error bars are 95% confidence bounds for the gradient fit.
Figure 10: Reference flame speed at different strain rates as a function of overall equivalence ratio $\phi_o$, for 9% (upper row) and 20% (bottom row) acetone droplet addition. Cases for $K = 0 \text{ s}^{-1}$, and the rich flames at $K = 100 \text{ s}^{-1}$ are obtained by linear extrapolation. The typical scatter for measurements is around 2 cm/s, as obtained from Figure 8. Data for gas phase is from [22].
Figure 11: Flame speed curves extracted at $K = 200 \text{ s}^{-1}$ for 0%, 9%, and 20% acetone. Both two-phase (4.7$\mu$m droplet) and gaseous reference cases are plotted.
Figure 12: A comparison between the gas-phase counterflow flame simulation (lines) using COSI-LAB and experimental data (markers) for (a) 0% acetone, (b) 9% acetone, (c) 20% acetone, and (d) pure acetone. The markers were extracted from the raw data of our previous work [22]. Each data point represents the mean reference flame speed averaged over 50 single-shots. Solid lines are based on the acetone/methane reaction mechanism proposed in [22]. In subfigures (b)-(d), simulation results based on another acetone mechanism introduced by Pichon et al. [47] are also plotted as dashed lines for comparison. The strain rates for the simulation range from 80-250 s⁻¹. Within this range, a linear behavior of flame speed to strain rate is supported by the simulation results.
Figure 13: Simulation results for (a) droplet diameter, (b) acetone molar fraction, (c) gas temperature, and (d) axial velocity profiles along the axial direction for 20% acetone, 4.7 µm droplet size at equivalence ratio 1.05 for different strain rates. The x-axis represents the axial distance from the nozzle (0 mm). All acetone was assumed to be in liquid form.
Figure 14: Evaporation curves for different initial acetone vapor fraction at the boundary. The strain rate is 149 s$^{-1}$. The $y$ axis is the molar fraction of vapor acetone in the mixture.
Figure 15: Comparison of experimental data and simulation results for (a) lean, (b) near stoichiometry, and (c) rich conditions for 20% acetone case. The gas simulations with a lower initial temperature are also conducted and plotted (green lines) for comparison with the droplet cases, so that the total enthalpy of the mixtures are equal. The initial temperature is calculated as 276 K, 274 K and 271 K for the three cases by subtracting the theoretical temperature drop from the room temperature (290 K). Note that for the gas data [22], the room temperature was 298 K.