Spray combustion in moderate and intense low-oxygen conditions

- An experimental study -

PROEFSCHRIFT

ter verkrijging van de graad van doctor
aan de Technische Universiteit Delft,
op gezag van de Rector Magnificus Prof. ir. K.C.A.M. Luyben,
voorzitter van het College voor Promoties,
in het openbaar te verdedigen
op donderdag, 22 januari, 2015 om 12.30 uur
door

Hugo Ricardo CORREIA RODRIGUES

diplom ingi, Faculty of Engineering of the University of Porto
geboren te Lamego, Portugal.
Dit proefschrift is goedgekeurd door de promotor:
Prof. dr. D.J.E.M. Roekaerts

Copromotor: Dr. ir. M.J. Tummers

Samenstelling promotiecommissie:

Rector Magnificus                       voorzitter
Prof. dr. D.J.E.M. Roekaerts           Technische Universiteit Delft
Dr. ir. M.J. Tummers                   Technische Universiteit Delft
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Prof. dr. ir. C.R. Kleijn (reserve)     Technische Universiteit Delft

This research was financially supported by the Technology Foundation STW (project 10418, part of the Clean Combustion Concepts Programme).

Printed by Ipskamp Drukkers B.V., Enschede, The Netherlands
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Author email: hugorcrodrigues@hotmail.com
To my parents...
Nitric oxides (NO\textsubscript{x}) are formed as a byproduct of combustion and contribute to ground-level ozone formation and the creation of conditions harmful for human health. In response to air pollutant emissions regulations, technologies for controlling NO\textsubscript{x} formation for an entire spectrum of fuels ranging from gaseous, liquid, coals, biomass to residue-derived fuel are of great importance. The technology of Moderate and Intense Low-oxygen Dilution (MILD) combustion (also known as HiTAC or FLOX®) holds the potential to maximize efficiency and reduce pollution emissions for liquid fuels. It relies on the dilution of recirculated combustion products in pre-heated air, yielding low temperature gradients within combustion chamber and NO\textsubscript{x} emissions.

This study concerns an experimental investigation of spray flames generated in a laboratory-scale burner designed for the combustion of liquid fuels in MILD conditions. The objective was twofold: 1) reach fundamental understanding of the process and, 2) create a database useful for validation of computational models. The successful validation of these models will aid the design and optimization of industrial combustion systems. The configuration used in this study consists of a spray jet injected into a coaxial upward flow of either air or hot-diluted combustion products. The latter case resembles an environment with temperature and oxygen dilution typical for advanced MILD combustors. An important characteristic of the developed system is that the properties of the fluid entrained in the near burner zone are controlled independently of the progress of combustion in the spray flame or the flue gas composition downstream of the flame. By contrast, in a typical furnace or boiler configuration, the properties of the entrained flow are determined by in-furnace aerodynamics and cannot be varied independently. The greater flexibility of this 'spray-in-hot-coflow' burner compared to a furnace or a boiler configuration, makes possible to study systematically a range of different conditions. The spray was created using a commercial pressure-swirl atomizer.
Ethanol and acetone were used as fuel because of their well-known physical properties and the availability of detailed and reduced chemical mechanisms for the combustion process.

Flames with several combinations of spray and coflow conditions were studied in detail, namely (1) ethanol reacting sprays in air and hot-diluted coflow, (2) ethanol reacting sprays in different hot-diluted coflow conditions, and (3) acetone and ethanol sprays in an identical hot-diluted coflow. High-speed visualizations of liquid breakup were performed to provide insight on the atomization mechanisms. Complementary laser-based diagnostic techniques, PDA and CARS were employed to characterise the properties of gas and liquid phase in the spray region. PDA provided simultaneous measurements of droplet velocity and size statistics and CARS the gas-phase temperature statistics. For the first time, CARS has been applied to determine gas-phase temperature statistics in a regions with high droplet density. The velocity and temperature statistics of the coflow were measured using respectively LDA and CARS. The composition of the coflow was measured using a flue gas analyser. The coflow measurements together with the measurements in the spray region as close as possible to the atomizer, provides a dataset of inflow boundary conditions useful for numerical simulations.

First, a comparative study of an ethanol spray in air and an ethanol spray in hot-diluted coflow was performed. These two test-cases serve as basis for comparison between conventional and MILD combustion of liquid fuels. The high-speed visualizations show that a liquid sheet cone emerges from the atomizer nozzle and disturbances, initiating from the nozzle tip, grow in space causing a local thinning of the liquid sheet and disruption. In the case of hot coflow, the presence of strong vaporization promotes the thinning of the liquid sheet and the onset of disruption takes place earlier. Although, in air and hot-diluted coflow differences are observed on the onset of disruption, a similar droplet size distribution in the near atomizer region is observed for both cases and the resulting differences in the spray flame structure farther downstream stem from the different properties of the entrained coflow, i.e. temperature and oxygen dilution. In the case of air coflow, an inner and an outer flame-front is observed. In the case of hot-diluted coflow, the heat-release in the inner flame-front is substantially smaller. A significant reduction of temperature samples above 2000 K is observed in the outer flame-front for the hot-diluted case.

Secondly, three ethanol sprays issuing in coflows with different temperature and oxygen dilutions were studied. The Weber number remains identical among the three cases and the temperature and the oxygen dilution were varied together. The results provide insights in the gas flow and the droplet distribution in the near atomizer field as well as the subsequent droplet dispersion.
Complementary to this set of measurements, a parametric study was made concerning the sensitivity of ethanol spray flames with the injection pressure (and, therefore, initial liquid jet velocity) as well as the coflow conditions. A linear relationship was found between the lift-off height and the injection pressure which is different from what has been observed in a similar burner for a gaseous fuel jet in hot coflow. The lift-off height was found to depend on the droplet convective, vaporization and chemical time scales prior to ignition. The spray flame structure does not change with the different coflow conditions. At the outer flame-front, the peak temperatures above 2000 K are found to correlate with the coflow conditions. Higher coflow temperatures (and, therefore, higher droplet vaporization rates) together with the lower oxygen dilution lead to an increase of the peak temperatures.

Thirdly, a comparative study was made of the structure of an ethanol and an acetone spray flame in identical coflow conditions. This study provides insight on the atomization mechanics, the stabilization processes and the resulting peak temperatures of liquid fuels with different physical properties burning in MILD conditions. Both flames are lifted but their visual characteristics are very different. The acetone case is found to have a higher lift-off height contrary to what would be expected based on comparison of the physical properties of the liquid fuels. High-speed visualizations shows that for acetone cavitation occurs inside the atomizer. As consequence, the initial droplet distribution, turbulent dispersion of droplets and turbulence modulation of the gas-phase by droplets changes drastically and this is also at the basis of the larger lift-off height of the acetone flame. The two flames also drastically differ in the nature of the spray in the outer region. In the ethanol flame, the outer region is predominantly filled by large droplets; in the acetone flame, small droplets with low inertia are present and quickly follow the mixing structures contributing to vaporization and the formation of an ignitable mixture.

The present study provides fundamental scientific knowledge and insight on the nature of MILD combustion for liquid fuels. In general it can be concluded that differences in properties of the recirculated hot combustion products yield different atomization characteristics and, subsequently, different turbulent dispersion and turbulence modulation of the gas-phase flow due to the presence of droplets. Although combustion in hot-diluted coflow conditions in general leads to absence of high peak temperatures, the degree in which peak temperatures are avoided depends on the interaction between coflow conditions and the characteristics of the atomization process. Additionally, the dataset offers new opportunities for model validation. Its value comes from the combination of several features such as: challenging complexity, degree of completeness of the dataset for each single case and availability of several cases with different
flame structure and liquid fuels.
Samenvatting

Stikstofoxide (NO\textsubscript{x}) worden gevormd als bijproduct van verbranding en dragen bij aan ozonvorming op grondniveau en creatie van condities die schadelijk zijn voor de menselijke gezondheid. Technologieontwikkeling in reactie op emissiewetgeving op het gebied van luchtverontreiniging zijn technologie en betreffende controle van NO\textsubscript{x} vorming voor een heel reeks brandstoffen, van gasvormig, vloeibaar, kool, biomassa tot zware olie is van uitermate groot belang. De technologie genoemd 'MILD combustion', (ook bekend als 'vlamloos', HiTAC of FLOX\textsuperscript{®}) heeft het potentieel om de efficiëntie te van de verbranding van vloeibare brandstoffen te maximaliseren en de emissie van vervuilende stoffen te verminderen. Ze steunt op het verdunnen van voorverwamde lucht met gerecirculeerde verbrandingproducten. Dit levert lagere temperatuurgradiënten op in de verbrandingskamer en lagere NO\textsubscript{x} emissions.

Deze studie betreft de ontwikkeling en de gedetailleerde studie van een laboratorium-schaal brandersysteem voor verbranding van vloeibare brandstof met behulp van MILD verbranding. Het objectief was tweevoudig: fundamenteel begrip bereiken van het proces en een database creëren die nuttig is voor het valideren van computationele modellen. Eens gevalideerd kunnen zulke modellen gebruikt worden als middel voor ontwerp en optimalisatie van industriële verbrandingsapparatuur. De configuratie bestaat uit een spray straal die geïnjecteerd wordt in een co-axiale stroming van ofwel lucht ofwel lucht verdund met hete verbrandingproducten. Het laatste geval lijkt wat betreft temperatuur en verdunning van het zuurstofniveau op de omgeving typisch voor MILD verbrandingskamers. De spray werd gecreëerd door gebruik te maken van een commerciële swirl-drukverstuiver. Ethanol en aceton werden als brandstof gebruikt omdat hun fysische eigenschappen welbekend zijn en er gedetailleerde en gereduceerde chemische reactieschema's beschikbaar zijn in de literatuur. Een belangrijke karakteristiek van het ontwikkelde systeem is dat de eigenschappen van het fluidum dat aerodynamisch ingezogen wordt in
het gebied nabij de brander kunnen gecontroleerd worden onafhankelijk van de voortgang van de verbranding in de spray vlam of van de samenstelling van het productgas stroomafwaarts van de vlam. Dit is in contrast met de situatie in een typische situatie in een formuis of boiler, waar de eigenschappen van de aangezogen stroming bepaald zijn door de aerodynamica in het formuis en niet onafhankelijk gevarieerd kunnen worden. De grotere flexibiliteit van deze 'spray-in-hete-cflow' brander in vergelijking met een formuis of boiler maakt het mogelijk op een efficiënte manier een studie te maken van de gevoeligheden bij een breed bereik aan condities.

Vlammen bij verschillende combinaties van spray en coflow werden in detail experimenteel bestudeerd, namelijk 1) reagerende ethanol sprays in lucht en in hete verdunte coflow, 2) reagerende ethanol sprays in verschillende hete verdunte coflow condities, en 3) acetone en ethanol sprays in een identieke hete verdunte coflow. Hoge-snelheid visualisaties van het opbreken van de vloeistof werden gedaan om inzicht te krijgen in de verstuivingsmechanismen. Complementaire laser-gebaseerde diagnostische technieken PDA en CARS werden gebruikt om de eigenschappen van de gasfase en de vloeistof fase in het spray-gebied te karakteriseren. PDA levert simultane metingen van de statistiek van druppelsnelheid en druppeldiameter en CARS levert de statistiek van de temperatuur van de gasfase. In dit werk is CARS voor het eerst toegepast om de temperatuur van de gasfase te bepalen in spray vlammen. De statistiek van snelheid en temperatuur van de coflow werd bepaald door gebruik te maken respectievelijk van LDA en CARS. De gemiddelde samenstelling van de coflow werd bepaald met een gasanalyse apparaat. De meting in de coflow en de metingen in de spray zo dicht mogelijk bij de verstuiver zijn van belang voor het bepalen van instroom-randvoorwaarden nodig in numerieke berekeningen.

Ten eerste werd een vergelijkende studie gemaakt van een ethanol spray in lucht en een ethanol spray in hete verdunte coflow. Deze twee test-systemen dienen als basis voor een vergelijking tussen conventionele en MILD verbranding van vloeibare brandstoffen. De hoge-snelheids visualisaties tonen aan dat er een kegelvormige vloeistoflaag uit de tip van de verstuiver komt en dat verstoringen die beginnen bij de nozzle tip groeien in omvang wat een locale verdunning van de vloeistoflaag veroorzaakt en disruptie. In het geval van de hete coflow versterkt de sterke verdamping het dunner worden van de vloeistoflaag en het begin van de disruptie vindt eerder plaats. Hoewel er verschillen worden waargenomen tussen de gevallen van lucht en hete verdunte coflow, blijkt er een in beide gevallen een gelijkardige druppeldiameterverdeling te zijn in het gebied nabij de verstuiver en de verschillen die optreden in de structuur van de spray vlam maken stroomafwaarts kloren van de verschillen in de eigenschappen van de aangezogen coflow, namelijk de temperatuur

Ten tweede werd een studie gemaakt van drie ethanol spras in coflows met verschillende temperatuur en zuurstofverdunning. Het Weber getal was identiek in de drie gevallen en de temperatuur en de zuurstofverdunning werden samen gevarieerd. De resultaten geven inzicht in de stroming van het gas en in de verdeling van druppels nabij de nozzle alsmede in de eropvolgende verspreiding van de druppels. Complementair aan deze set metingen werd een parametrische studie gemaakt van de gevoeligheid van de ethanol sprayvlam voor injectiefuik (en dus initiële vloeistofstraal snelheid en voor de coflow condities. Een lineair verband werd gevonden tussen de hoogte waarop de vlam stabiliseert (lift-off hoogte) en de injectiedruk. Dit verschilt van wat geobserveerd werd in het geval van een gelijkaardige brander met een gasvormige jet in hete coflow. De lift-off hoogte bleek af te hangen van de tijdschalen voor convectie verdamping en chemische reactie voorafgaand aan ontsteking. De structuur van de spray vlam verandert niet met de verschillen in coflow condities. De temperaturen boven2000 K in het buiten-vlamfront blijken te correleren met de coflow condities. Hogere coflow temperatuur (en daarom ook hogere verdampingssnelheid van druppels) samen met lagere zuurstofverdunning leiden samen tot een toename van de piektemperaturen. Ten derde werd een vergelijkende studie gemaakt van vlammen van een ethanol en een aceton spray in identieke coflow condities. Deze studie leverde inzicht in de verstuivingsmechanismes, de stabilisatieprocessen en de resulterende piektemperaturen voor vloeibare brandstoffen met verschillende fysische eigenschappen bij verbranding in MILD condities. Beide vlammen zijn los van de brandertip maar hun karakteristiek visueel uitzicht is erg verschillend. In het geval van aceton blijkt de lift-off hoogte hoger te zijn, wat tegengesteld is aan wat verwacht zou worden enkel en alleen op basis van de fysische eigenschappen van beide vloeibare brandstoffen. Hoge-snelheid visualisaties tonen aan dat voor aceton cavitation optreedt in de verstuiver. Als gevolg hiervan veranderen de initiële druppelverdeling, de turbulente verspreiding van druppels en de modulatie van de turbulentie in de gasfase door de druppels drastisch. En dat is ook de basis voor de grotere lift-off hoogte van de aceton vlam. De twee vlammen verschillen ook drastisch wat betreft de aard van de spray in het buitengebied. In de ethanol vlam wordt het buitengebied voornamelijk bereikt door grote druppels; in de aceton vlam zijn er daar kleine druppels aanwezig met een kleine
inertie die snel de mengstrukturen volgen wat bijdraagt aan een significante verdampering en warmtevrijgave binnen de beschikbare tijd.

In het algemeen kan er geconcludeerd worden dat verschillen in de eigenschappen van de gerecirculeerde verbrandingsproducten verschillende verstuivingskarakteristieken opleveren, en in vervolg daarop, verschillende turbulente verspreiding van de druppels en verschillende turbulentiemodulatie van de gasfase door de aanwezigheid van druppels. Hoewel verbranding in een hete verdonde coflow conditie in het algemeen leidt to afwezigheid van hoge piektemperaturen en de corresponderende lage NO\textsubscript{x} vorming, is de mate waarin piektemperaturen vermeden worden afhankelijk van de interactie tussen coflow condities en de karakteristieken van het verstuivingsproces. Deze studie levert een waardevolle database op voor modelvalidatie. Haar waarde komt voort uit de combinatie van verschillende eigenschappen: uitdagende complexiteit, mate van volledigheid van de dataset voor elk bestudeerd geval en beschikbaarheid van meerdere gevallen met verschillende vlamstructuren en vloeibare brandstoffen. Deze studie levert fundamenteel wetenschappelijke kennis op en inzicht in de aard van MILD verbranding van vloeibare brandstoffen.
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CHAPTER 1

Introduction*

1.1 Energy and low NO\textsubscript{x} combustion technologies - Current state

Oil provides 35% of our current total energy consumption, and more oil is used today than at any other time. Daily oil consumption in 2012 stood at about 90 million barrels per day (an increase of about 15% over 2000 levels) [2]. This rapid growth is forecast to continue, with consumption predicted to reach 120 million barrels per day by 2030 [2]. Prompted by the rapidly rising fuel consumption and increased emissions of carbon dioxide (CO\textsubscript{2}) and nitrogen oxides (NO\textsubscript{x}) into the environment, it is now a matter of great urgency to deal with environmental preservation on a global scale.

To meet these challenges the search goes on for new combustion strategies beyond what may be called 'conventional combustion'. In Fig. 1.1 a schematic of conventional combustion and a novel combustion process named Moderate and Intense Low-oxygen Dilution (MILD) is presented.

In conventional air combustion the fuel and air oxidizer streams are injected separately and combustion occurs after the fuel and the oxidizer mix on the molecular scale. The combustion is associated with the occurrence of a complex sequence of exothermic chemical reactions. During this process, combustion products are generated and accompanied by the release of heat and radiation [68]. Nearly all flames are visible to the human eye and are, therefore, called luminous. The luminous emission arises from: 1) the radiative emission from combustion gases within the flame that emit in the luminous region of the spectrum and 2) almost pure carbon particles (called soot) which are generated during the combustion process. Additionally to the luminous emission, a significant portion of radiation is emitted in the infrared mainly by

*The contents of this chapter have been published in:
soot particles as well as the carbon dioxide and water vapor. Since the radiation depends on the absolute temperature to fourth power order, the greater the temperatures, the stronger the thermal radiation and, consequently, the more luminous the flame becomes. Thermal radiation becomes an important heat transfer mechanism with rising temperature levels and may be totally dominant over conduction and convection at very high temperatures. Thus, combustion strategies that maximize heat transfer without incurring penalties in pollutant formation and emissions are of great importance.

In combustion systems using hydrocarbon fuels, carbon dioxide is always present in the combustion products and it is proportional to the carbon content of the fuel, unless it is artificially removed [25]. If a suitable recovery of the heat enthalpy of the exhaust gases is organized, and the gained energy is used, the carbon dioxide emission is reduced for the same thermal input power of the combustion system. Additionally to carbon dioxide, nitric oxides formation, i.e. nitric oxide (NO) and nitrogen dioxide (NO₂), may occur through the reaction of molecular nitrogen, contained in fuel or from the combustion air, with oxygen. When emitted to the atmosphere, those affect significantly the
ozone balance and contribute to the undesirable formation of photochemical smog [45, p. 409-491]. There are predominantly three sources of nitric oxides from combustion processes, prompt NO\textsubscript{x}, fuel NO\textsubscript{x} and thermal NO\textsubscript{x} [94]. The formation of thermal NO\textsubscript{x}, described by the 'Zel'dovich-mechanism', is based primarily on the temperature and oxygen concentration. Therefore, any reduction of combustion gas temperature or availability of oxygen can produce low nitric oxide emissions.

Different techniques can be employed to reduce combustion temperature [49]. Some techniques rely on controlling combustion temperature by the amount of excess air premixed in the gaseous fuel. This technique is of special interest for gas turbines applications, however, an intrinsic feature of premixed-fuel combustion systems is a tendency toward flashback [24, p. 353-365]. Flashback occurs when the flame travels upstream from the combustion zone into the premixing sections of the combustor and can result in several safety issues. Other techniques rely on the withdrawal of energy from the flame (e.g. flame cooling and staging). However, this results give rise to problems associated with flame stability and incomplete combustion products such as carbon monoxide. In certain cases and depending on the industrial application, combustion air may be preheated. In this way, the issue of flame stability is overcome and the thermal efficiency of the combustion system increased by capturing part of the energy in the exhaust gases. However, this strategy results in a increase of peak flame temperatures and higher thermal efficiencies are achieved at the cost of higher nitric oxides production.

The technology of Moderate and Intense Low-oxygen Dilution (MILD) combustion allows to reduce thermal nitric oxides together with an increase of the thermal efficiency [3, 41, 49]. The operation principle of this technology, also referred to as Flameless Oxidation (FLOX\textsuperscript{®}), Excess Enthalpy Combustion (EEC) or High Temperature Air Combustion (HiTAC), is displayed in Fig. 1.1. Combustion air is preheated above the auto-ignition temperature of the fuel by recovering partially the energy contained in the exhaust gases. Prior to the mixing with the fuel, the pre-heated air is blended with the recirculated combustion products in the combustion chamber resulting in a significant reduction of the local oxygen concentration by increasing the inert content of the combustion mixture. Thus, the combustion reaction is impeded, leading to a distributed reaction zone which occupies a larger volume than that of 'conventional combustion'. This, in turn, has the effect of creating a more uniform temperature distribution over the length of the flame distributing the reaction over a larger volume. Furthermore, the local peak temperatures are reduced while maintaining the total heat production.

A theoretical analysis based on a canonical diffusion flame can be used
to describe the processes that characterize MILD conditions and illustrate its essential features (see Fig. 1.2). Consider a system with two separate fuel (subscript 1) and oxidizer (subscript 2) streams in the gas state, it is possible to quantify the degree of mixing by a mixture fraction variable that represents the chemical elements locally available. In the limit of infinitely fast irreversible reactions and assuming equal diffusivity of all species i.e. Lewis (Le) number equal to one, the adiabatic temperature \( T_{ad} \) as a function of the mixture fraction can be defined as [75]:

\[
T_{ad} = T_{amb} + Q Y_{F,1} \frac{Z_{st}}{1-Z_{st}}, \quad Z \geq Z_{st} \tag{1.1}
\]

\[
T_{ad} = T_{M}(Z) + \frac{Q Y_{O_2,2}}{c_p} Z, \quad Z \leq Z_{st} \tag{1.2}
\]

\[
Z_{st} = \left[ 1 + \frac{s Y_{F,1}}{Y_{O_2,2}} \right]^{-1} \tag{1.3}
\]

where \( T_{M} = Z T_1 + (1-Z) T_2 \) is the temperature after non-reactive mixing, \( Q \) is the heat released per kg fuel and \( s \) is the oxidiser to fuel mass ratio for complete combustion.

Figure 1.2: Illustration of the Burke-Schumann solution as a function of mixture fraction for conventional and MILD combustion for the same fuel. \( T_{ig}, T_{M} \) and \( T_{oxy} \) refer to ignition, mixture and hot oxidizer temperature respectively.
1.2 Motivation and Objectives

Fig. 1.2 illustrates the temperature profiles as a function of the mixture fraction variable in a conventional and MILD environment. The equations are known as Burke-Schumann solutions. Conceptually, equations (1.1) and (1.2) show that a reduction of the oxygen concentration \( Y_{O_2} \) in the oxidizer stream, (which is achieved by the increase of the inert content in the mixture) leads to a reduction in the flame adiabatic temperature and, therefore, hampers thermal nitric oxides formation. Additionally, as show by equation (1.3), it results in a shift of the peak adiabatic temperature towards leaner mixtures. This strategy, however, leads to practical problems associated with the flame stability since the heat-release is proportional to the local amount of oxygen available and the chemical process can not self-sustain. When the temperature of the oxidizer stream is elevated above auto-ignition temperature, the thermal energy needed to sustain and promote the reaction is already contained in one of the reactants. Thus, the lean mixtures resulting from the molecular process of diffusion and the bulk convective motion, can react even when the enthalpy released in the reaction is a small fraction of the total. In this way the thermal nitric oxides formation is minimized.

1.2 Motivation and Objectives

A search of the literature shows the success of MILD combustion concepts for industrial scale furnaces operating with various fuels [49, 85]. However, large scale furnaces design precludes any quantitative experimentation. The goal of this study is to provide an experimental database in a laboratory-scale burner with easy optical access for laser-based diagnostics. The results will be used for modeling and numerical validation and provide a valuable insight into the design of a boiler operating in MILD conditions with heavy-fuel oils.

Fuel oils are derived from crude oil which is a mixture of hydrocarbons and small amounts of nitrogen, sulfur, and vanadium (see Fig. 1.3a). The fraction of each of the aforementioned components varies according to the place of origin.

Fuel oils can be classified by the components resulting from the distillation process. They are divided in two major types [58]: Distillate fuel oils and Residual fuel oils. *Distillate fuel oils* (see Fig. 1.3b) are vaporized and condensed during a distillation process. When the fractional distillation process is applied to crude oil, the gaseous and light substance boil off first, followed by gasoline, kerosene, and then light and heavy distillate fuel oils (gas oils). This petroleum fractions have a definite boiling range and do not contain high-boiling oils or asphaltic components. *Residual fuel oils* (also known as heavy-fuel oil) are
the residual material from the distillation process and is obtained after other lighter petroleum products such as gasoline and kerosene have been distilled off. Residual fuel oil is a black fuel composed mostly of carbon, hydrogen, sulphur and other impurities such as ash, metals, and water (see Fig. 1.3c). Since crude oil composition varies according to the petroleum source, there is a considerable variability in the composition of residual oils.

Alternatively, for commercial purposes fuel oils can be classified according to its physical properties and the industrial applications for which they are suitable. Standard D396 [1] divides fuel oils into six classes, numbered 1 through 6, according to its boiling point, composition and purpose. The boiling point, ranging from 175 to 600°C, and carbon chain length, 20 to 70 atoms, of the fuel increases with fuel oil number. Viscosity also increases with number, and the heaviest oil has to be heated to get it to flow. Price usually decreases as the fuel number increases. Grade 1 and 2 oils are commonly referred to as light-fuel oils or domestic fuels and include kerosene, stove oil, and furnace fuel oil. These are distillate fuel oils used primarily in applications that do not require atomization by air or steam in order to reduce droplet size for proper burning such as domestic heating boilers and furnaces. Grade 3 is a distillate fuel oil and is rarely used. Grade 4 is usually a blend of distillate and residual fuel oils used in commercial/industrial burners equipped for this viscosity range. Grade 4 may be 100% residual material. However, for practical purposes, it is generally blended with sufficient distillate stock to meet viscosity and flash point requirements for the grade. Grade 5 and 6 are called residual fuel oils.

Figure 1.3: Images of distillate fuel oil and residual fuel oil samples.
1.3 Turbulent multiphase combustion in MILD conditions

(or heavy fuel oils) used in industrial burners. Preheating is usually required for handling and proper atomization.

Although higher grade oils are low-cost fuels with relevant energy content (e.g. heating value of about 35 and 38 MJ/kg for grade 5 and 6, respectively), the burning of higher grade fuels has been target of severe control emissions legislations due to the high pollutant emissions such as NO\textsubscript{x}. In view of its relevant energy content and respond to the growing requirement of many combustion systems for multifuel capability, coming to an understanding of the physics of MILD combustion regime for liquid fuels is essential.

1.3 What is specific about turbulent multiphase combustion in MILD conditions?

Liquid fuels typically are delivered into the combustion chamber as turbulent sprays so that the fuel can effectively burn. Turbulent spray flames are essentially two-phase reacting flows with a continuous- (gas) and a dispersed-phase (droplets). Thus, a great deal can be learnt from turbulent gas flames. Gaseous flames, for either laminar or turbulent flows, can be categorized in nonpremixed, premixed, or partially premixed depending on the degree of premixing prior to combustion [75]. At the global level, gaseous flames consists of two reactants: a fuel and an oxidizer. When injected separately, these two are brought together through the molecular process of diffusion and the bulk convective motion, to a common region where mixing and subsequently reaction takes place. These flames are labeled as nonpremixed flames. In a premixed system, an ignitable mixture is already present in the reactants stream. Diffusion is still needed to transport the heat from the reaction zone to the cold flammable mixture.

Turbulent spray flames differ from gaseous flames in that the initial fuel liquid jet first desintegrates into ligaments and droplets (see Fig. 1.4). These initial ligaments and droplets, depending on the aerodynamics forces exerted, proceed into further breakup until the surface tension overcomes the aerodynamics forces. These two stages of fragmentation are referred in the literature as primary and secondary atomization [35, 74]. Atomization of a liquid fuel represents a key aspect in combustion efficiency and performance of practical combustion systems since the disintegration of a liquid jet into droplets promotes fuel evaporation and combustion by generating a very high surface-to-volume ratio in the liquid phase. Different atomizers, often tailored to specific combustor geometries, produce different patterns and the resulting droplet
distribution are mainly predicted by empirical models based on the atomizer geometry and on the liquid relative velocity and physical properties. In the case of MILD combustion the oxidizer is hot and strong vaporization is expected to occur early in the near-atomizer region and to change the atomization process. To the author knowledge, no theory that accounts for the strong vaporization effects in the physical mechanisms at work in the atomization process is available in the literature.

Farther downstream, droplets are aerodynamically stable and secondary breakup ceases to exist. This region, referred to as 'dilute-region', is characterized by lower droplet concentration and spherical droplets with larger inter-droplet distance as compared to the dense-region. A first challenge arises from the relative rates of droplet vaporization versus oxidizer availability surrounding the droplets. Specifically, the liquid fuel is sprayed into the hot oxidizing gas that initially has little or virtually no oxidizer present within the spray. Since the hot-diluted oxidizer gas has to be entrained into the jet interior for reaction to be possible, the spray combustion characteristics then depend critically on the relative rates of droplet vaporization versus available oxidizer surrounding the droplets. Furthermore, once ignition takes place, droplets are known to evaporate and burn in single or group modes, interacting with one another [9, 52, 53, 54, 66]. A cluster of works dealing primarily with single droplet combustion in hot oxygen reduced environment show that a depletion of the oxygen available in the surrounding environment of a single-droplet leads to an increase of the flame radius and, therefore, group combustion modes and strong interaction between the flames are to be expected in MILD conditions.
The second challenge is the strong coupling of fluid dynamics and chemical kinetics over wide ranges of spatial and temporal scales (see Fig. 1.5). The interaction of turbulence and chemistry produces temporal and spatial fluctuations in the mixing rates influencing the flame stability, relation among species concentrations and, ultimately, the formation of pollutants. The relative importance of the rates of chemical reaction and fluid dynamic mixing is characterized by the Damköhler number (Da). Conceptually, Da → 0 represents a chemically frozen situation where the reaction time is excessively long relative to the characteristics flow time available for the reaction to proceed. The other extreme represents a situation of fast-chemistry where reaction is completed instantly. In MILD conditions the dilution of the exhaust gases with reduced oxygen concentration, slows down chemical reaction rates [64]. The result is that the characteristic time of kinetics and turbulence are comparable (Da → 1) and the two are strongly coupled with each other.

\[
Da = \frac{\text{Characteristic flow time scale}}{\text{Characteristic chemical time scale}}
\]  

(1.4)

Figure 1.5: Illustration of the chemical-physical time scales interaction in turbulent multiphase reacting flows (extracted from Warnatz et al. [48]).
The droplet dispersion into the gaseous phase brings additional complexity to the process physics. The presence of relative velocities between droplets and the gaseous phase contributes to enhanced droplet evaporation rates and, subsequently, to more fuel vapor available in the continuous phase. The mixing of the strong turbulent recirculated gases with the resulting fuel vapor from the droplets contributes to a decrease of the fine fuel concentration gradients. The turbulence structure of the continuous-phase is known to be modulated by the presence of the dispersed-phase since the relative motion between the fluctuating velocities of the dispersed and continuous-phase provides an additional mechanism for turbulent energy dissipation [5, 20]. This alteration may, in turn, depending on the local droplet mass loading, manifest itself has significant changes in the Damköhler number. Hence, the local mixture fraction and the continuous-phase properties are strongly dictated by the droplets dynamics.

1.4 Laboratory-scale setups and modelling challenges

The science of two-phase reacting flows involves intricate intercoupling between the phenomena observed in gaseous flames and droplets. During the last three decades, computer simulations have been used in the analysis and design of engineered systems. However, development and validation of advanced combustion models that can then be used to develop cleaner, more efficient combustion systems are needed.

The International Workshop on Measurement and Computation of Turbulent Nonpremixed Flames (TNF) is an ongoing series of meetings focused on the investigation of the turbulence-chemistry interaction for turbulent nonpremixed and partially premixed combustion [88, 89]. It is an excellent example of collaborative experimental-numerical work with the aim to establish a library of well-documented target flames that are appropriate for model validation as well as the advancement of basic scientific understanding of turbulent combustion.

Examples of target flames used for model development are presented in Fig. 1.6. The flames have been selected to cover a progression in complexity, starting with simple jet flames of H$_2$, H$_2$/He, and H$_2$/N$_2$ in air. Dilution with N$_2$ or He reduces flame temperature, minimizing the influence of radiation on NO formation and better isolating the effects of turbulence-chemistry interaction. Because these hydrogen flames are fully burning (attached to the
1.4 Laboratory-scale setups and modelling challenges

Figure 1.6: Target flames examples: (a) CO/H$_2$/N$_2$ jet flame, (b) CH$_4$/H$_2$/N$_2$ jet flame, (c) piloted CH$_4$/air jet flame, (d) piloted natural gas jet flame, (e) CH$_4$/H$_2$ bluff-body flame, (f) CH$_4$/air swirl flame, (g) lifted H$_2$/N$_2$ flame in vitiated coflow, (h) CH$_4$/air turbulent opposed jet flame, (i) TECFLAM non-premixed flame, (j) enclosed lifted CH$_4$ swirl flame.

nozzle and without local extinction) and kinetically relatively simple, they may be addressed using a broad range of models. Jet flames of CO/H$_2$/N$_2$ and CH$_4$/H$_2$/N$_2$ increase the chemical kinetic complexity, while maintaining a simple flow field.

Similarly, the International Workshop on the Turbulent Combustion of Sprays (TCS) reports on the progress of experimental and numerical techniques in two-phase flows, with emphasis on spray combustion [17, 18]. The motivation for studies in this area is that knowledge of the dominant phenomena and their interactions in such flow systems is essential for the development of predictive models and their use in combustor and gas turbine design. This
necessitates the development of accurate experimental methods and numerical modelling techniques of reactive multiphase flows.

![Figure 1.7: Target flames examples: (a) CH\textsubscript{4}O hollow cone spray flame, (b) C\textsubscript{2}H\textsubscript{6}O pressure-swirl spray flame, (c) piloted C\textsubscript{2}H\textsubscript{6}O dilute spray flame, (d) lifted C\textsubscript{2}H\textsubscript{6}O dilute spray flame in hot-diluted coflow.](image)

A complete overview of available experimental databases and model validation studies of reactive multiphase flows is presented in the work of Jenny et al. [73]. In Fig. 1.7, some examples of experiments used as benchmark studies to cover different aspects of reactive multiphase flows are shown. Regarding the level of coupling between the flow field and the remaining physics (e.g. chemical reactions, heat transfer), the level of complexity of the benchmark spray flames is not extensive as in large scale combustion systems. Specifically, the laboratory-scale burners are simplified and designed to couple with complementary laser diagnostics and other measurement techniques. This allows to meet accuracy requirements for model validation as well as provide well-described boundary conditions. Also, in view of the validity of the droplet evaporation models and the computational capabilities required for numerical simulations, only single-component fuels with well-documented reduced chemical mechanisms are selected. Complex fuels like heavy-fuel oils contain many components and the evaporation of corresponding fuel droplets cannot be accurately described using an effective single component representation [87]. If the fuel is a multi-component mixture, components with lowest molar mass tend to evaporate most quickly and experience a different mixing history than the heavier components. As a consequence, ignition and flame structure may be sensitive to the multi-component nature of the fuel which, in the case of heavy-fuel oils, depend significantly on the place of origin. Additionally, disruptive boiling or microexplosions are also possible since for certain regions
of the droplet interior, the equilibrium vapor pressure of the more volatile component can exceed the ambient pressure [23, 98].

Widmann and Presser [50] measured the properties of a hollow-cone turbulent methanol spray enclosed in a combustion chamber. The NIST experimental facility (a) was developed to permit well-defined input and boundary conditions, enabling measurements to characterize the fuel spray, combustion air, wall temperatures, gas temperatures, and species concentrations. Heidelberg spray flame (b) was designed to generate a stable flame without the use of a bluff body or a pilot flame facilitating at some extend the numerical simulations. A turbulent ethanol spray flame was characterized through quantitative experiments using laser-based techniques. The experiments include spatially-resolved measurements of droplet sizes (Mie/LIF-dropsizing and PDA), droplet velocity (PDA), liquid-phase temperature (2-color LIF temperature imaging with Rhodamine B) and gas-phase temperature (multi-line NO-LIF temperature imaging). As shown in Fig. 1.7, it was demonstrated that different flame structures could be achieved by adjusting the fuel pressure and the coflow conditions and, thus, present challenges regarding the formation mechanism of the spray flame structures.

Numerical results on the NIST (a) and Heidelberg case (b) show that droplet size measurements at the inlet profile causes uncertainties in the spray and gas flow results and may result in different flow fields. To circumvent the issue of liquid disintegration and the inlet conditions in the near-injector region, other studies have been conducted in laboratory-scale burners using nebulizers instead of atomizers [14, 92, 99]. A nebulizer can disperse modest liquid flowrates while imparting minimal velocity to the liquid drops originating the typical environment of a spray dilute-region. Stårner and Masri [92] studied turbulent piloted dilute spray flames (c) using an ultrasonic nebulizer mounted upstream of the jet exit plane and air and nitrogen as carrier gas. The piloted jet burner produces a simple streaming flow of droplets and carrier gases and uses a set of premixed flames using a stoichiometric mixture of hydrogen, acetylene and air to stabilise the main jet to the burner’s exit plane. OH/acetone laser-induced fluorescence images reveal a substantial similarity to premixed flame behavior when the carrier stream is air. When the carrier is nitrogen, the reaction zone has a diffusion flame structure.

O’Loughlin and Masri [99] extended the burner design to study the effect of a hot-diluted coflow on a dilute spray. The burner facility consists of a dilute spray, i.e. droplets and carrier issuing in a coflow of hot combustion products. Droplet loading on the carrier gas and the coflow conditions were systematically changed. The velocity field data and droplet fluxes are provided but the composition data are limited to planar imaging of selected species.
Simultaneous high-speed OH-CH* Planar LIF and droplet Mie scattering show that ignition OH kernels formation and growth is the mechanism of spray flame stabilization in hot-diluted conditions. This brought new challenges and opportunities for model validation concerning the stabilization mechanisms of spray flames.

The goal of this study was to design a burner facility capable of producing turbulent spray flames in MILD conditions and, ultimately, be used for model validations. As in the NIST and Heidelberg cases, it was opted to use a commercial pressure-swirl nozzle to atomize the liquid fuel. Based on literature survey and expertise at Delft University of Technology three aspects were considered of great importance: a) develop a burner facility with simple flow geometry and well-defined inlet conditions, b) use of single-component fuels with well-described reduced chemical mechanisms and, c) obtain sufficiently wide range of data for each test case to ensure reliable modeling. The last, concerns measurements of the inflow conditions and several variables at different relevant locations. Complementary laser-based techniques and high-speed visualizations were carried out to obtain a comprehensive set of detailed measurements in the near-injector region and farther downstream. To the author knowledge, for the first time gas-phase temperature statistics were measured in regions with high droplet density providing new opportunities for model validation. Also, datasets on different cases showing trends and changes in combustion regimes were obtained.

1.5 Outline of the thesis

In this thesis, ethanol and acetone sprays in MILD conditions are examined by complementary pointwise laser techniques and high-speed visualizations. This thesis is mainly a compilation of published and submitted papers (Chapter 3-5). Thus, to highlight the common parts among the papers, a detailed description of the laboratory-scale burner and the measurements techniques is presented in Chapter 2. The pointwise laser-based measurement techniques employed in this study are: laser Doppler anemometry, phase Doppler anemometry and coherent anti-Stokes Raman spectroscopy. High-speed visualizations were performed with a CCD camera to examine the liquid fuel disintegration into droplets. Flue-gas analyser was employed to measure the $O_2$ distribution across the hot-diluted coflow. Measurement positions are briefly presented on the cases description section of each chapter.

In Chapter 3, a study of ethanol spray flames in air and hot-diluted coflow is presented. The main goal is to unveil the flame structure originated from
a pressure-swirl spray in 'conventional' and MILD conditions. A discussion of the visual features observed in the liquid disintegration is given. The subsequent droplet dispersion into the coflow and temperature statistics in the reaction zones are discussed. In Chapter 4, the effects of different hot-diluted coflow conditions in the spray flames structure are studied. The coflow temperature and oxygen dilution are varied together. A discussion of the overall visual appearance of the flames is presented. The lift-off behaviour is shown to be largely dependent on the droplet convective and vaporization time scales prior to ignition. The droplet dispersion and the temperature statistics across the spray flame are also discussed. In Chapter 5, the investigations are extended to acetone liquid fuel. The qualitative features of liquid breakup for ethanol and acetone are presented. A comparison of the spray flame structure for both fuels in the same hot-diluted coflow conditions is given. The temperatures statistics are also presented. Finally, main conclusions and recommendations are presented in Chapter 6.
This chapter focused on two main aspects: 1) the design criteria and working principle of the burner facility, 2) the description of the measurement techniques used in the present study. The burner consists of a spray issuing in an upward coflow with well-controlled inlet conditions. Measurements of the size and velocity of the droplets, as well as of gas-phase velocity and temperature were made by Phase Doppler Anemometry (PDA) and Coherent Anti-Stokes Raman Spectroscopy (CARS), respectively. The coflow inlet conditions were described by means of Laser Doppler Anemometry (LDA). Time resolved highspeed visualizations were performed to qualitatively analyse the breakup of different liquid fuels.

2.1 Burner design criteria

The principal role of the coflow is to deliver a hot-diluted oxidizer with an bulk temperature and oxygen representative of MILD conditions. The following list of design requirements are key factors in the collaborations of numerical and experimental research efforts and results from the literature survey and expertise at Delft University of Technology.

- The spray and coflow configuration will be axisymmetric and therefore may be amenable to 2D modeling.
- The secondary burner shall be compatible with seeding particles used as flow tracers for laser diagnostics.
- The coflow diameter and velocity should be large enough to isolate the jet flame from the laboratory air for the maximum downstream distance possible.
- The coflow stream should be stable and uniform.
- The control system should provide a range of coflow capabilities (e.g.
range of coflow temperature, stoichiometry and velocity).

- The liquid fuel used shall have well-documented physical properties and kinetically simple.

2.2 Description of the burner facility

In Fig. 2.1, pictures of the burner facility are presented. The schematic of the burner facility is displayed in Fig. 2.2. The burner consists of a pressure-swirl atomizer that produces a spray of fine fuel droplets issuing in a coflow of either air or hot combustion products. Hereafter, these two cases will be referred to as 'air coflow' and 'hot-diluted coflow' respectively.

The air is introduced into the burner through four inlets and straightened by a stack of perforated plates yielding a balanced flow with homogenous and isotropic turbulence prior to the secondary burner. The hot-diluted coflow is produced by the secondary burner that operates on air and Dutch Natural Gas (DNG). The typical molar composition of DNG is 81.3% methane, 14.4%
2.2 Description of the burner facility

Figure 2.2: Schematic of the burner facility with relevant dimensions in millimeters. At the right-inside, the top view and a plane cut (A-A) along the secondary burner are presented.

nitrogen, 3.7% ethane and 0.6% of trace species. The secondary burner consists of an hexagonal packing arrangement of identical 5 mm diameter vertical pipes with each end welded to perforated plates. At half-height of each individual vertical pipe, four 0.5 mm holes were cut in a cross-configuration. The secondary burner is sealed with six wedges cut to the size and clamped into place by screws. The working principle is as follows: DNG is injected evenly through each of the six wedges inlets and is forced out into the vertical pipes through the cut 0.5 mm holes. As the incoming air flows through the vertical pipes, mixing occurs and, eventually, yields a matrix of flamelets. The vertical round-pipe downstream of the secondary burner establishes a heat-loss through radiation to the surroundings, lowering the initial coflow temperature.
Experimental setup

to a nominal value that mimics the practical environment of burners operating in MILD conditions. Also, it prevents the coflow from mixing with the surrounding laboratory air throughout this process, preserving the established coflow oxygen concentration.

The burner design presents two limitations: 1) hot-diluted coflow temperature and oxygen concentration cannot be varied independently. Hence, the air-to-DNG ratio together with the effects of two perforated plates and the pipe length dictates the hot-diluted coflow temperature, oxygen concentration and turbulence levels. Dilution of air with N\textsubscript{2} prior to the secondary burner can be used to overcome this issue. However, so far such has not been implemented and the experiments are limited to the range of temperature achieved without dilution. 2) Depending on the air and DNG mass flow rates selected a strong 'humming' can be observed. This phenomena causes large pressure fluctuations and yields an unsteady behavior and distortion of the spray flame. The undesirable tonal rumble was eliminated by a trial and error approach. 'Humming' originates from the coupling between the coflow temperature, the pipe length and, ultimately, the axial position of the perforated-plates along the vertical pipe. The procedure included changing the coflow operating temperature, pipe length and the axial location of the perforated plates and the respective inter-distance. It was found that for a vertical pipe length of 280 mm, no 'humming' is heard within a coflow temperature and oxygen dilution range typical of MILD conditions.

Above a certain axial location the effects of the entrainment of the surrounding air into the hot-diluted coflow have substantial effects on the flame structure [78, 79]. These flames are essentially a three-stream mixing problem, i.e. fuel jet, hot-diluted coflow and surrounding air, increasing to a great extent the complexity of data interpretation and analysis as well as of numerical simulations [37, 61]. Hence, the coflow diameter was chosen large enough to isolate the spray flame from the surrounding air as much as possible. The axial location at which the perturbation of the laboratory air takes place is presented in the following chapters.

The spray was produced by a commercial pressure-swirl solid cone atomizer (Delavan WDA 0.5 GPH) with an 0.21 mm exit orifice and a 60° spray angle mounted on a 2 mm inner diameter stainless steel pipe through which liquid fuel is fed to the atomizer. To prevent excessive heating and possible pre-vaporization of the liquid fuel, the fuel pipe is water-cooled and several layers of thin stainless steel foils wrapped around the cooled fuel pipe to form a heat shield. Liquid fuel temperature is monitored at the atomizer inlet by means of a type K thermocouple (Thermocoax). Pressure is measured with a pressure transducer (Omegadyne MMA). The analog output of the pressure
transducer is fed to the external data collection system of a TSI PDA system for simultaneous measurement of pressure and droplet size and velocity. The burner can move in three orthogonal directions by means of a traversing system equipped with computer-controlled stepping motors. Throughout the thesis a r, z-coordinate system will be used with its origin at the atomizer orifice exit plane. The z-coordinate is measured along the axis of the burner and ‘r’ denotes the radial coordinate.

Figure 2.3: Block diagram of the components comprising the burner facility.

In Fig. 2.3 a diagram of the burner facility is presented in a block outline. Several components are used to achieve stable and accurate mass flow rates for air, DNG and liquid fuel. The air line consists of a filter, pressure regulator, cyclone seeder and Bronkhorst mass flow controller with an inaccuracy of ±0.5% of the full scale. The cyclone seeder was loaded with alumina oxide particles (Al₂O₃) and only used for LDA measurements.

The available pressure in the local distribution line of DNG is insufficient to reach the required mass flow rates and, therefore, a compressor was used. To avoid the undesirable pressure fluctuations induced by the compressor, a pressure dampener and a pressure regulator was used prior to Bronkhorst mass flow controller. The Bronkhorst mass flow controller selected has also an inaccuracy of ±0.5% of the full scale. The two liquid fuels used in this study were ethanol (C₂H₅OH) and acetone (CH₃COCH₃). The liquid fuel was stored into a vessel at ambient pressure. A pump was used to raise the liquid fuel to
a desirable pressure and the mass flow rates monitored by a Bronkhorst mini-Coriolis mass flow controller with ±0.2% inaccuracy. The local water supply was employed to: 1) cool to the laser systems and, 2) run the water-cooling system of the burner. Water was circulated through copper coils immersed in a water bath in a chiller unit. The water bath temperature was monitored throughout the experimental campaigns by a thermometer.

2.3 Measurement Techniques

To adequately understand each of the measurement techniques, a brief explanation of the working principles is given. This section provides an overview of the physics related to each of the individual measurement techniques used in the present study as well as system layout and data processing considerations. The reader is referred to Albrecht et al. [42] and Eckbreth [11] for further details.

2.3.1 Laser Doppler Anemometry (LDA)

Physical background

![Figure 2.4: Illustration of the measurement volume fringe pattern and signal generation.](image)

The simplified drawings presented in Fig. 2.4 can help to understand the operation principle of LDA. The wavefronts of two coherent beams interfere
in the intersection volume, forming interference fringes with a spacing \(d_f\). The interference pattern consists of alternating zones of brightness and darkness, i.e. of high and low intensity energy levels. The beams exhibit a Gaussian intensity profile in its cross section and, therefore, the fringes at the center have higher intensities than those at the border. As a particle moves through the fringes, scattered light fluctuates in intensity with a frequency equal to the velocity of the particle divided by the fringe spacing. The photodetector records a signal burst whose amplitude is modulated by the fringe pattern. The frequency of the modulation is the Doppler frequency, \(f_D = \frac{2 V_p}{\lambda} \sin(\theta/2)\), where \(V_p\) is the velocity component perpendicular to the fringes.

Depending on the relative position of the photodetector to the measurement volume, the system collects the scattered light either in forward or back-scattering. The main advantage of a back-scattering layout is that both transmitting and receiving optics are integrated in same optical system and, therefore, no individual alignment of the beams is required.

An identical modulated signal will be obtained for particles with identical velocity magnitude and opposite direction. This problem is obviated by Bragg shifting the beams \(f_s\), so the fringes are moving relative to the flow. Thus, a tracer particle with zero velocity in the measurement volume will result in a received frequency of \(f_s\), while any movement of the particle will result in a frequency larger or smaller than \(f_s\), depending on the direction relative to the fringe movement.

### LDA system layout

LDA measurements were performed with a two-component, dual beam TSI-system operating in back-scattering mode. A illustration of the components of the system is presented in Fig. 2.5. The green line (514.5 nm) and blue line (488 nm) of a 10 W Continuum Argon-ion laser were used to measure axial and radial velocity components \(U_z\) and \(U_r\), respectively. A Bragg cell generates the required frequency shift between both beams so as to eliminate directional ambiguity of the particles. The transmitting optics (3) had a focal length of 350 mm and yielded a measurement volume of 0.17 mm in diameter and 2.40 mm in length. The photomultiplier output signals (5) were fed to a TSI FSA-3000 processor (6). Alumina oxide particles \((\text{Al}_2\text{O}_3)\) with an average diameter of 1 \(\mu\)m were added to the air by using a cyclone seeder. At each measurement position \(10^4\) samples were acquired to ensure adequate convergence of the second-order moments. Velocity statistics were computed using the transit time as a weighting factor to compensate for the velocity bias [42]. Consequently, the velocity statistics should be interpreted as Favre-averaged
2.3.2 Phase Doppler Anemometry (PDA)

Physical background

PDA additionally to the velocity statistics, brings forth the possibility to include the size information of spherical droplets. This laser-based technique is based on the spatial frequency of a scattered interference pattern. Fig. 2.6 presents a schematic of the PDA operation principle. As in LDA, two intersecting beams with a Gaussian laser beam intensity form an interference pattern in the measurement volume. The receiving optics differs from LDA in that the photodetectors are placed at a certain off-axis collection angle ($\phi$) measured against the direction in the plane of the laser beams. When a spherical droplet passes through the fringes of the measurement volume, it scatters light.

The light scattered by a spherical droplet in a plane wave front has contributions from reflection at the external surface of the droplet, refraction through the droplet and those refracted after one and two reflections at the inner surface of the droplet. The receiving lens projects a portion of the fringe quantities [31].

Figure 2.5: Illustration of the LDA system layout and respective components.
pattern onto three detectors and each detector produces a Doppler burst signal with a frequency proportional to the particle velocity (see Fig. 2.7).

Considering that 1) the droplets have larger diameters than the wavelength of light and 2) only single scattering mode is present, by geometrical optics it is possible to establish a relation between the phase shift of the Doppler burst signals from two different detectors and the size of the droplets. For
Experimental setup

Figure 2.7: Theoretical phase-diameter relations obtained by the geometrical optics approximation.

first-order refraction the relation between the droplet diameter and the phase shift ($\beta$) between detector 1 and 2 is given by:

$$\Delta \beta_{12} = \frac{2\pi d_d m \sin \Psi \sin \theta/2}{\nu \sqrt{1 + m^2 - m \nu}}$$

Eq. (2.1) states that the phase difference varies linearly with the droplet diameter. The determination of the phase-diameter relations then requires the refractive index and the angle between the three detectors. As shown in Fig. 2.7, the refractive index and the PDA configuration parameters influence the system sensitivity and are of importance to achieve accurate results. In a reacting spray, the temperature increase of the droplet and its surrounding gaseous phase results in a reduction of the relative refractive index. Thus, the measurements of droplet diameter are subject to bias if the droplets properties are not measured simultaneously. In the spray flame in air coflow case ($A_{II}$) presented in Chapter 3, at the lift-off height axial position, the refractive index ranges from 1.365 to 1.333 over the temperature range from 20°C to near boiling at 78.1°C. This results in a systematic error up to 3% in the droplet diameter. More downstream in the spray where droplets are close to boiling, the effect of temperature on the refractive index of ethanol is negligible. For the spray flames in hot-diluted an estimation of the droplet temperature at the first measurement station indicates that droplets are at boiling temperature. Hence, this effect does not occur.
PDA system layout

PDA system provides simultaneous measurements of droplet velocity and size statistics in the spray region. A representative schematic of the PDA system used in the present study is laid out in Fig. 2.8. The transmitting optics (3) used for the PDA are identical to those of the LDA system. The receiving optics (4) were set up to collect mainly first-order refracted light (p=1). The front and internal lenses had focal lengths of 650 mm and 250 mm, respectively. The measurement volume with size $0.17 \times 2.4 \text{ mm}^2$ was imaged onto a 150 µm slit. The scattered light was detected by photomultipliers in a photo Detector Module (6). The signal processing was performed by a TSI FSA-4000 processor (7) and the associated FlowSizer software (8). At each measurement position $3 \times 10^4$ samples were acquired except at those with data rates less than 100 Hz where $10^4$ samples were taken.
**Data Processing and practical considerations**

A spray consists of a polydispersion of droplets sizes and care was taken to obtain a correct measurement of all aspects. Specifically, quantities such as droplet representative diameters and volume fluxes depend on the instrument settings such as Argon-ion output power, gain used for the detectors, filter size and threshold levels. A parametric study of the adjustable operational parameters influence in the droplet diameter, droplet number density and velocity was performed in a position within a dilute region of the spray in hot-diluted coflow. The volume fluxes depend on these three quantities and it is possible, due to error propagation that, each individual quantity is measured incorrectly and yet accurate flux values are obtained. In view of this, volume fluxes were not taken in consideration. The optimization procedure was as follow: The filter size on the green and blue channels was first set according to the observed range of velocities in the dense spray. The laser output power and photomultiplier gain were increased independently until changes smaller than 0.2% were observed. No appreciable differences were observed with the threshold levels and the signal-to-noise ratio.

An intensity validation scheme from TSI software was used to account for multiple droplets, trajectory-dependent scattering errors and reject large drops that are erroneously sized as small drops. The parameters of the intensity validation scheme were determined in a very dilute region of the spray flame. The established parameters were kept constant throughout the measurement campaigns. The inherent phase difference introduced by electronic circuits on the acquisition data was calibrated using the procedure proposed by TSI [95]. The Gaussian light intensity distribution in the measurement volume introduces a bias into the droplet size statistics. Specifically, larger droplets scatter more light and therefore can be detected farther away from the center of the probe volume than smaller droplets. A probe volume correction for the droplet size statistics was used based on the maximum path length for each particle size class.

The droplet density varies throughout the spray domain and the number of samples collected shall be sufficiently high to achieve the convergence of the statistical moments within a feasible measurement time. A total number of \(10^4\) samples were collected at positions with data rates lower than \(10^2\) Hz. In all the remaining positions, ensembles of \(3 \times 10^4\) samples were acquired. The physical dimensions of the measurement volume formed by the intersection of the two laser beams is a limiting parameter in that only a single droplet passing through a measurement volume can be measured [55]. Hence, the number density measurements in the region near the atomizer is inherently
2.3 Measurement Techniques

underestimated.

The study of the gas motion and the relative droplet motion in two-phase flows relies on the identification of small droplets that follow faithfully the gas phase. The selection of the upper size limit of droplets was estimated by computing the gas-phase statistics with several droplet thresholds. An additional aspect taken in consideration is that Lorenz-Mie oscillations of the phase/diameter curve are relevant for droplets smaller than 10 µm since the geometrical optics is not a good approximation of the Mie theory for very small particles and regions with low signal-to-noise ratio such as the spray dense region. Thus, only droplets with a ±1 µm tolerance between the phase difference from the three-detectors were considered from the full records.

2.3.3 Coherent Anti-Stokes Raman Spectroscopy (CARS)

The pioneering work in developing CARS for combustion diagnostics was carried out during the 1970's by Taran and co-workers [36, 82, 83]. Since then, the technique has been developed to the point of its being accepted as a standard for temperature measurements in high interference environments typical of practical combustion processes. Substantial reviews of CARS and of its progress can be found in [10, 11] and the references therein. In the present research, the potential of CARS to provide reliable gas-phase temperature measurements in the regions with high density liquid droplets was explored. The background information needed to understand its working principle will be presented first. The discussion will be followed by the practical considerations and the specific issues concerning its implementation in spray flames.

Background physics

Fig. 2.9 displays a schematic representation of the CARS optical layout and the energy level diagram of the photon coupling for an isolated transition in a gas mixture. The CARS signal is generated by means of a four-wave-mixing nonlinear optical process. When the frequency difference from the incident pump and Stokes laser beams (ωp - ωs) matches the frequency of a particular Raman active rovibrational transition (ωv,J), the electric field polarizes a dielectric medium through a displacement of the electron cloud relative to the atomic nuclei. The photons from the probe laser beam (ωpr) are then scattered from the induced states, producing a strong anti-Stokes signal at ωas = ωpr + (ωp - ωs) that completes the four-wave mixing process. The anti-Stokes signal that is emitted includes the induced Raman shift and, therefore, is spectrally shifted 'towards the blue' relative to the green probe
beam. In order to observe the several Raman shifts for all populated levels in one single laser shot, a broadband Stokes laser is typically used. This leads to the entire CARS spectrum with each pulse permitting instantaneous measurements of medium properties. The Raman shifts are dependent upon the specific energy levels of the probed molecule and, therefore, the spectral information serves as a fingerprint of the molecule state and, ultimately, on the gas temperature within the probe volume.

To better understand the CARS signal dependence on the transient molecular response as well as the effects of the field laser sources, a mathematical formulation of the CARS signal is first presented. The equation for the CARS signal strength is derived by means of the electromagnetic wave equation obtained by manipulating Maxwell’s equations. The details of deriving of the CARS signal equation are not shown here, but can be found in the comprehensive work of Boyd [90]. It leads to a final expression of:

\[
I_{AS} = \frac{\omega_{AS}^2}{n_p^2 n_s n_{AS} c^4 \varepsilon_0} I_p^2 I_s |\chi_{CARS}^{(3)}|^2 l^2 \sin^2 \left( \frac{\Delta k l}{2} \right)^2
\]  

(2.2)

Where \(I_P, I_S, I_{AS}\) are the beam intensities at frequencies \(\omega_p, \omega_S\) and \(\omega_{AS}\), respectively. \(|\chi_{CARS}^{(3)}|\) is the third-order susceptibility, \(l\) is the distance of the
2.3 Measurement Techniques

probe volume and $sinc^2$ term the phase matching condition. As it follows from Eq. (2.2), the dependencies upon the strength of the signal are those of the laser intensity, phase matching condition and the third-order susceptibility the last having a much smaller magnitude compared to the other terms. The strength of the CARS signal is linearly dependent upon the intensity of each of the generating beams and, therefore, the use of high-power lasers is favourable for generating a strong signal. However, an upper threshold for the energy beam exists. Above a certain energy level, dielectric breakdown occurs and the bright plasma discharge produces a background signal that obscures the CARS signal. The third-order susceptibility can be written as follows for a single photon transition:

$$\chi^{(3)}_{\text{CARS}} = \frac{8\pi^2 n_p e_0 c^4 N \Delta N_{\nu,J}}{n_s \hbar \omega_s^4 \left( \omega_{\nu,J} - \omega_p + \omega_s - j \frac{\Gamma_{\nu,J}}{2} \right)} \left( \frac{\partial \sigma}{\partial \Omega} \right)_{\nu,J}$$ (2.3)

where $N$ is the number of molecules, $\Delta N_{\nu,J}$ the fractional population difference between the states involved in the wave mixing, $\omega_{\nu,J}$ the Raman shift, $\Gamma_{\nu,J}$ the linewidth and $\partial \sigma/\partial \Omega$ the differential Raman cross section. The population fractions of rovibrational states are given by the Boltzmann equation:

$$N_{\nu,J} = \frac{g_J (2J + 1)}{Q_{\text{vib}} Q_{\text{rot}}} \left[ e^{\frac{G_{\nu,J}}{kT_{\text{vib}}}} - e^{\frac{-F_{\nu,J}}{kT_{\text{rot}}}} \right]$$ (2.4)

with $g_J$ representing the nuclear spin multiplicity and $Q_i$ the partition function.

Three major inferences can be drawn from Eq. (2.3) and (2.4). First, the exponential dependence of $N_{\nu,J}$ on the temperature gives rovibrational spectroscopy its sensitivity to temperature. Specifically, the CARS spectrum consists of a number of spectral lines and its shape and its position are highly temperature dependent. The key attribute of thermometric sensitivity of the technique is thus based on the temperature-dependent amplitude of the $J$ specific spectral lines originating mainly from the population difference factor as indicated in Eq. (2.3). Secondly, the CARS signal is coherent in nature and emitted in a specific solid angle. Thus, as compared with other techniques such as spontaneous Raman scattering, stronger laser-like signals are obtained. Third, $\chi_{\text{CARS}}$ can be divided into a resonant and a non-resonant part. As it will be shown later, the presence of droplets along the beam path yield additional contributions to the measured spectrum and special care was taken to correct for this effect.
CARS experimental setup

Figure 2.10: Illustration of the CARS system and respective components.

A representative schematic of the CARS system is presented in Fig. 2.10. The ro-vibrational CARS system is based on an injection-seeded, frequency-doubled Nd:YAG laser (1) (Spectrun SL805 SLM), which yields 500 mJ per pulse at 532 nm with a pulse duration of 12 ns at 10 Hz repetition rate. About 80% of the radiation is used to pump a modeless Stokes dye laser (3) (Mode-X ML-3), emitting a broadband profile around 607 nm for Rhodamine 640 in methanol in order to probe the N\textsubscript{2} molecules. The remaining 20% of the pump laser travels along a delay line and is split into two beams with equal intensity. The Stokes beam and the two beams at 532 nm are focused by an aplanat lens (5) with a focal length of 300 mm in a planar BOXCARS scheme yielding a CARS probe volume of 700 µm length and 35 µm diameter. During the experimental campaigns, significant radiation is emitted from the burner to
its surroundings. To avoid instabilities in the laser power output and optical elements, the lasers and part of the optics were located in a separate room.

The generated CARS radiation is recollimated and separated from the 532 nm beam. By means of an echelle spectrometer (13), the CARS spectrum is dispersed on a CCD detector with $1100 \times 330$ pixels. The binned spectra is digitized by an 18-bit converter and stored.

**Data Processing and practical considerations**

The spectrum is subtracted from the background and normalized using a non-resonant spectrum measured from methane. Dacapo software is used to fit the spectrum to a library of theoretical, temperature-dependent spectra, convoluted for the instrument broadening. The temperature obtained by the best fit between measured and calculated spectrum is found by an iterative method. Previous experiments show that single-shot imprecision of the system is 1-4% over a range from 2000 K to 300 K. The inaccuracy is estimated to be 20 K [33, 34].

Results of the fitting procedure are displayed in Fig. 2.11. The distinct single-shot imprecision at flame and room temperature results from the number of spectral lines for the evaluation. In Fig. 2.11, an example of the measured spectrum and corresponding fitted spectrum in the work reported in Chapter 3 is displayed. One of the single-shot spectra refers to a position in air coflow (Fig. 2.11a) and other in the flame region (Fig. 2.11b). At room temperature, $N_2$ vibrates mainly with the ground level frequency and the spectrum is an unresolved band of Q-branch transitions ($\nu = 0 \rightarrow 1, \Delta J = 0$) originating from the low rotational levels. Thus, slight distortions in the measured spectrum may have greater impact on the estimated temperature. With increasing temperatures, excited states start to become thermally populated and transitions from higher vibrational and rotational levels are observed which spread out in energy for increasing J. As a consequence, individual Q-branch transitions become visible.

Two considerations were taken into account in the CARS measurements: one being the impact of the droplets on the resonant $N_2$-CARS signal, the other the effects of strong temperature gradients within the CARS probe volume. Two situations can be distinguished depending on the relative position of the droplet with respect to the probe volume. In the presence of a droplet in the probe volume, a dielectric breakdown is observed, disrupting or obscuring the resonant $N_2$-CARS signal and, consequently, no valid CARS temperature measurements is obtained. In the event of a droplet along the laser beam paths, an incidental increase in the background occurs. The increase con-
Figure 2.11: Results of a CARS spectrum fitting procedure.

sists of a sloping signal, sometimes accompanied with broad structures (see Fig. 2.12). Usually, a linear spectral background profile could be evaluated by interpolation between values found at the left-and right-hand sides of the N$_2$-CARS spectrum. A steep temperature gradient may occur over the probe
volume, particularly near the reaction zone of the flame. In the presence of such a gradient, a substantially lower temperature than the mean temperature will be calculated due to the density effect. Such was corrected based on a weighted fit according to the procedure suggested by [33].
2.3.4 High-speed Visualization

The liquid jet breakup was recorded by using an imaging system consisting of a light source and a high-speed camera. To back-light the liquid jet a powerful LED light (Waldmann - MCXFL 3S) was set on-axis with the camera lens. Images of the liquid breakup region were captured using a Photron Fastcam SA1 camera. A lens bellow (Nikon PB-6) was coupled to the camera allowing the lens (Micro-Nikkor AF 200mm) to be moved relative to the chip plane and, hence, reduce the field of view.

2.3.5 Flue gas Analyser

Oxygen volume fraction measurements were performed with a Testo 335 flue-gas analyser, with a specified inaccuracy of ±0.20%.
A laboratory scale experimental setup was built to study ethanol pressure-swirl spray flames in a coflow of either air or hot-diluted oxidant. The latter case resembles conditions similar to those found in practical combustion systems of liquid fuels operating in Mild conditions. First, experiments have been performed to investigate the phenomena associated with the atomization process. High-speed visualizations show that in the presence of a hot-diluted coflow, an almost immediate disruption of the liquid jet takes place, indicating significant changes in the atomization mechanism, compared to the case with air coflow. Secondly, a comprehensive set of measurements was obtained by complementary single-point measurement techniques to reveal the gas and droplets flow fields as well as temperature fields. Measurements of droplet size and velocity components in the spray region were obtained by Phase Doppler Anemometry. Gas temperature was measured using coherent anti-Stokes Raman Spectroscopy even in regions with droplet density as high as $10^5 \text{cm}^{-3}$. It has been observed that in a reacting spray in air coflow, an inner and an outer flame-front are present. For a reacting spray with similar injection pressure in hot-diluted coflow, weakening of the inner flame-front is attributed to the fact that the gaseous mixture becomes increasingly rich towards the center region. Consequently, a significant reduction of occurrence of temperature samples above 2000 K is observed throughout most of the spray region.

*The results of this chapter have partially been published in:

3.1 Introduction

The technology of MILD combustion, or 'flameless combustion' relies on the dilution of fresh reactants with cooled recirculated combustion products prior to the main combustion zone thereby lowering flame adiabatic temperature and, consequently, reducing significantly nitric oxides ($NO_x$) emissions [49]. For this process to be effective, a combination of sufficiently large recycle ratio of combustion products and a mixture temperature exceeding the auto-ignition temperature of the fuel is required [3, 16, 62].

The most notable and comprehensive experimental studies on MILD conditions were performed in laboratory scale non-premixed gaseous flames. Dally et al. [19] developed a jet-in-hot-coflow (JHC) burner and performed measurements of reactive scalars in a $CH_4/H_2$ flame. The oxygen mass percentage in the oxidizer coflow ranged from 3% to 9%. Modeling studies of these flames revealed that the accurate representation of the initial coflow radial profiles of velocity statistics and scalar statistics are of utmost importance [61]. Oldenhof et al. [30, 31] constructed a similar JHC burner, but modified the secondary burner to allow optical measurement techniques for flow velocity based on tracer particles. Mean velocity and temperature fields were measured in non-premixed DNG flames in a hot coflow with oxygen mass percentage ranging from 7.6 to 10.9% [31]. A small temperature increase was found in the shear region and, velocity and turbulent stresses profiles in the reaction zones were not significantly affected by the chemical reactions [31]. High-speed recordings of the luminescence at the flame base, showed that the stabilization mechanism is based on auto-ignition kernels that grow into large pockets [30].

Compared to turbulent gaseous flames, in turbulent spray flames additional phenomena come into play. Mixing of a desintegrating liquid jet with its surroundings differs from that of a gaseous jet. To create the spray flame, the fuel stream first has to be desintegrated into an ensemble of droplets with a desirable distribution of sizes and velocities in order to achieve the required rate of vaporization, chemical heat-release, levels of conversion and pollutant emissions. Instabilities at the liquid-gas interface in the near-atomizer region lead to primary and secondary break-up. The liquid fragments in surrounding gas form a dispersed multiphase flow, where droplets are evaporating and subject to turbulent dispersion. The dispersed droplets modify the gas-phase turbulence and the released fuel vapor depending on the relative speed of evaporation, mixing and combustion, burns in diffusion flames around individual droplets or clusters of droplets, or in partially premixed gaseous flames [73]. Also the temperature and composition of the oxidizer influences the time scales
of phase change and reaction, making a spray flame in hot-diluted combustion regime different from that of a conventional spray flame in air.

The viability of combustion of liquid fuel in a diluted oxidizer at industrial and semi-industrial scale has been investigated in the literature. Tsuji et al. [41] reported experiments in a kerosene spray flame oxidised by preheated air diluted with combustion products with an oxygen percentage ranging from 3% to 10%. Weber et al. [85] performed experiments on MILD combustion of light fuel oil in a furnace at a power of 0.58 MW. Recently, Reddy et al. [96, 97] reported experimental and numerical studies of a two-stage labscale combustor where MILD conditions were reached using intense swirl. These investigations provide encouraging results on the applicability of MILD combustion to liquid fuels. However, the configurations studied did not allow for detailed laser diagnostic measurements needed to obtain insights on the fundamental aspects of this combustion technique.

The present work concerns an experimental study. The objective is to reach understanding, in an analogous way as done for gaseous fuels, by using a special burner design allowing for detailed observations. A laboratory scale burner was developed to study spray combustion under MILD conditions. The burner design retains the relevant physical processes of practical combustion systems. However, the composition of the gases entrained into the spray are controlled by a secondary burner rather than the furnace aerodynamics. Moreover, the spray flame is unconfined, allowing good optical access for complementary laser diagnostic techniques. Ethanol was used as fuel because of the well-defined properties. Fuel atomization is done using a commercial pressure-swirl atomizer. To clearly identify the characteristics of MILD combustion in comparison to conventional combustion two cases are considered. In the first case the fuel jet is injected in a coflow of ambient air and in the second case in a hot-diluted coflow with 9.2% oxygen by volume. This study contributes to the current literature in three ways: a) by advancing the use of laser diagnostic techniques in turbulent spray flames. This is done by reporting, to the authors’ knowledge, for the first time gas-phase temperature statistics in the spray region by means of coherent anti-Stokes Raman spectroscopy technique; b) by providing a comprehensive set of measurements that can be further used as valuable database for model validation of polydisperse sprays [73]; c) by providing understanding of the qualitative features of the transition of a liquid jet into a mixture of vapor and stable droplets in a hot surrounding environment.

It is known that the liquid atomization process and the near-atomizer region plays a key role in the entire spray flame characteristics and dynamics also further downstream [17, 39]. Advanced diagnostic techniques in two-phase
flows have been introduced at a slower pace than the gaseous counterparts and it has been difficult to obtain sufficiently accurate experimental data in near field of spray flames. To circumvent this problem, in the literature fundamental studies of turbulent spray flames have been made using a nebuliser instead of a pressure atomizer. In this configuration the complexity induced by the near-atomizer effects in the dense region of a pressure atomizer can be avoided. Karpetis and Gomez [12] reported on a series of experiments using methanol fuel. O’ Loughlin and Masri [99] extended the burner design of Karpetis and Gomez to study the effect of a hot-diluted coflow on a spray. Simultaneous high-speed OH-CH* Planar LIF and droplet Mie scattering show that ignition OH kernels formation and growth is the mechanism of spray flame stabilization in hot-diluted conditions. It may be argued though that the oxygen mole fraction in the coflow (12% by volume) is rather high compared to other studies of combustion systems operating in MILD conditions [41, 60].

In the present study a commercial atomizer was used for generating the spray instead of a nebulizer, because we considered it important to remain close to the most common configuration used in industrial practice. All measurements are made up to locations as close to the dense phase region as possible. The use of new experimental techniques such as ballistic imaging [63] and X-Ray absorption methods [102], would enable further progress towards a more complete measurements. Also detailed numerical studies [47] have the potential to provide understanding and detailed description of the atomization process. These advanced experimental and numerical techniques could be used to complement the observations using high speed camera reported here.

This paper is structured as follows: Section 3.2 presents the input parameters for three test-cases: non-reacting evaporating spray, reacting spray in air coflow and reacting spray in hot-diluted coflow. Section 3.3 is divided in three parts: 1) Comprehensive description of the coflow inlet turbulent characteristics; 2) high-speed visualization and analysis of liquid jet breakup and, last, 3) a discussion of the most notable features of spray flames structure in air and hot-diluted coflow. In Section 3.5, conclusions are presented.

### 3.2 Cases description

Table 3.1 summarizes the characteristics of the three different cases NA\(_I\), A\(_II\) and H\(_II\) that were considered in this study. Case NA\(_I\) corresponds to a non-reacting spray in air coflow and was included to elucidate the different features of the liquid sheet breakup by means of a high-speed visualization. Case A\(_II\) refers to a reacting spray in air coflow whereas H\(_II\) refers to a reacting spray...
3.2 Cases description

in hot-diluted coflow. Detailed measurements were carried out only for cases AII and HII and a summary of the experimental techniques and measurements locations are presented in Table 3.2. Results of other cases with different degree of coflow dilution will be report elsewhere.

Table 3.1: Spray and coflow input parameters for the studied test cases.

<table>
<thead>
<tr>
<th>Test cases</th>
<th>NA</th>
<th>AII</th>
<th>HII</th>
</tr>
</thead>
<tbody>
<tr>
<td>( \dot{m}_F ) [kg/h]</td>
<td>0.70</td>
<td>1.70</td>
<td>1.46</td>
</tr>
<tr>
<td>( p ) [bar]</td>
<td>5.1</td>
<td>12.3</td>
<td>11.5</td>
</tr>
<tr>
<td>( T_F ) [K]</td>
<td>293</td>
<td>296</td>
<td>301</td>
</tr>
<tr>
<td>We [-]</td>
<td>0.46</td>
<td>3.26</td>
<td>0.46</td>
</tr>
<tr>
<td>( \dot{m}_{air} ) [kg/h]</td>
<td>70</td>
<td>20</td>
<td>51</td>
</tr>
<tr>
<td>( \dot{m}_{DNG} ) [kg/h]</td>
<td>-</td>
<td>-</td>
<td>2.13</td>
</tr>
<tr>
<td>( U_{cf} ) [m/s]</td>
<td>0.820</td>
<td>0.245</td>
<td>2.243</td>
</tr>
<tr>
<td>( T_{cf} ) [K]</td>
<td>293</td>
<td>293</td>
<td>1200</td>
</tr>
<tr>
<td>( \langle X_{O_2} \rangle ) [%]</td>
<td>21</td>
<td>21</td>
<td>9.2</td>
</tr>
</tbody>
</table>

Table 3.2: Summary of measurements locations for AII and HII test cases.

<table>
<thead>
<tr>
<th>Measurement technique</th>
<th>( z ) (mm)</th>
<th>Comment:</th>
</tr>
</thead>
<tbody>
<tr>
<td>LDA</td>
<td>0, 20, 25, 30, 35, 40, 45, 50</td>
<td>Coflow radial profiles, Spray center axis</td>
</tr>
<tr>
<td>CARS</td>
<td>0, 15, 20, 30, 40, 45, 50, 60</td>
<td>Coflow radial profiles, Spray region</td>
</tr>
<tr>
<td>Flue gas analyser</td>
<td>0</td>
<td>Coflow radial profiles</td>
</tr>
<tr>
<td>PDA</td>
<td>8, 10, 12, 15, 20, 30, 35, 40, 45</td>
<td>Spray region, Steps of 1 mm</td>
</tr>
</tbody>
</table>

The quantities \( p \) and \( T_F \) represent the measured ethanol pressure and temperature, respectively. \( U_{cf}, T_{cf} \) and \( \langle X_{O_2} \rangle \) represent the coflow bulk velocity, temperature and oxygen volume fraction. For AII and HII, these quantities
Conventional and hot-diluted combustion regime

were obtained by integrating the measured profiles over the cross sectional area of the coflow inlet \((z = 0 \text{ mm})\). For case NA, the coflow bulk velocity is determined from the measured air mass flow rate. The Weber number \((\text{We})\) is defined as \(\rho U_j^2 l/\sigma\). \(l\) is the characteristic liquid film thickness \((l)\) and it was assumed as equal to the nozzle diameter, i.e. 210 \(\mu\text{m}\). \(U_j\) represents the difference between the estimated liquid jet velocity from the liquid mass flow rate and the coflow bulk velocity.

The main focus of the paper is on the comparison between the cases A and H. In case H significant heat transfer towards the central rod takes place thereby slightly increasing the liquid fuel temperature and, consequently, changing both viscosity and surface tension. This manifests itself as an increase of the injection pressure for the same liquid mass flow rate as compared to the corresponding air coflow case. Since a reduction in liquid mass flow rate is observed to lead to a loss of symmetry of the spray flame, it was opted to study spray flames with sufficiently high injection pressure. The change in fuel thermal input from case A to H is about 13%.

3.3 Results and discussion

3.3.1 Visual appearance of the spray flames

To show the overall flame appearance, color photos of the spray flames for cases A and H are shown in Fig. 3.1. The right inset shows a close-up of the spray region and initial development of the flame. The field-of-view measures 220 \(\times\) 170 \(\text{mm}^2\) (width \(\times\) height) and is the same in both cases. The photos were acquired with the same camera settings. In both cases, the atomizer yields a conical spray flame having an enlarging diameter downstream. The images show that flame A has a stronger luminescence and is shorter than H.

Three regions can be distinguished in the reacting spray in air coflow case (A) from its visual appearance: a jet liquid breakup region with no apparent presence of reaction, a bluish reaction zone and a sooty yellow zone far downstream. Lift-off height is defined here as the axial distance from the atomizer orifice exit plane, \(z = 0 \text{ mm}\), to the lowest axial position of the bluish reaction zone. Above the lift-off location, two flame-fronts develop from the flame base, one on the coflow side and another inside the spray. Such structure has been previously observed in methanol spray flames surrounded by an annular air duct [4, 28]. Three regions can also be distinguished based on visual appearance for the reacting spray in hot-diluted coflow (H): a jet liquid
breakup region, a faint bluish reaction zone and faint yellowish zone farther downstream. However, compared to case A_{II}, the flame is weakly luminescent, more transparent and relatively bright in the outer-regions of the spray.

### 3.3.2 Coflow details

LDA, CARS and flue gas analyzer were separately used to provide information on velocity, temperature and gas composition at the coflow inlet ($z = 0$ mm). The mean axial velocity $\langle U_z \rangle$ and turbulence intensity $u'_z/\langle U_z \rangle$ are plotted in Fig. 3.2 for case A_{II}. Fig. 3.2 also shows the results of additional measurements for a situation without fuel supply to the atomizer to illustrate the effect of the spray on the coflow inlet conditions. The coflow mean axial velocity $\langle U_z \rangle$ and the turbulence intensity $u'_z/\langle U_z \rangle$ for case H_{II} are presented in Fig. 3.3. A summary of the turbulence statistics and derived quantities at two radial positions in the coflow is given in Table 3.3 for cases A_{II}, H_{II} and in the case of an air coflow without the spray. The degree of anisotropy of the coflow is characterized by the ratio between the root-mean-square values of the axial and radial velocity fluctuations $u'_z/u'_r$. The Taylor microscale $\lambda_g$ was determined from autocorrelation functions constructed from time series of the axial velocity component with $2.5 \times 10^5$ samples by using the slotting method with local normalisation [67]. Taylor’s hypothesis was used to convert timescales into length scales. An estimation of the kinematic viscosity is required to compute the Taylor-scale Reynolds numbers. For case H_{II}, this was done by using $\langle X_{O_2} \rangle$ and $\langle T \rangle$ at the respective radial position as input parameter in detailed equilibrium chemistry calculations with Chemkin-Pro software [86].
Figure 3.1: Images of test cases with 0.8s exposure time and identical aperture. The field-of-view of the inset has dimensions $220 \times 170\,\text{mm}^2$ (width $\times$ height).
3.3 Results and discussion

Figure 3.2: Mean axial velocity $\langle U_z \rangle$ and turbulence intensity $u'_z / \langle U_z \rangle$ for case $A_{II}$ at $z = 0$ mm.

Figure 3.3: Mean axial velocity $\langle U_z \rangle$ and turbulence intensity $u'_z / \langle U_z \rangle$ for case $H_{II}$ at $z = 0$ mm.
Table 3.3: Summary of the relevant turbulence statistics in the coflow.

<table>
<thead>
<tr>
<th></th>
<th>w/o spray</th>
<th>A&lt;sub&gt;II&lt;/sub&gt;</th>
<th>H&lt;sub&gt;II&lt;/sub&gt;</th>
</tr>
</thead>
<tbody>
<tr>
<td>Radial position</td>
<td>R [mm]</td>
<td>19 50</td>
<td>12 50</td>
</tr>
<tr>
<td>Axial velocity</td>
<td>⟨U&lt;sub&gt;z&lt;/sub⟩ [×10&lt;sup&gt;2&lt;/sup&gt;] [m/s]</td>
<td>18.16 28.94</td>
<td>93.05 23.64</td>
</tr>
<tr>
<td>Radial velocity</td>
<td>⟨U&lt;sub&gt;r&lt;/sub⟩ [×10&lt;sup&gt;2&lt;/sup&gt;] [m/s]</td>
<td>3.05 1.82</td>
<td>65.55 6.57</td>
</tr>
<tr>
<td>Axial turbulence intensity</td>
<td>u&lt;sub&gt;z&lt;/sub&gt;′/⟨U&lt;sub&gt;z&lt;/sub⟩⟩ [%]</td>
<td>18.16 2.68</td>
<td>2.30 3.37</td>
</tr>
<tr>
<td>Axial r.m.s. of fluctuations</td>
<td>u&lt;sub&gt;z&lt;/sub&gt;′ [×10&lt;sup&gt;2&lt;/sup&gt;] [m/s]</td>
<td>6.32 0.77</td>
<td>2.14 0.79</td>
</tr>
<tr>
<td>Radial r.m.s. of fluctuations</td>
<td>u&lt;sub&gt;r&lt;/sub&gt;′ [×10&lt;sup&gt;2&lt;/sup&gt;] [m/s]</td>
<td>4.60 0.81</td>
<td>1.92 1.04</td>
</tr>
<tr>
<td>Anisotropy</td>
<td>u&lt;sub&gt;z&lt;/sub&gt;′/u&lt;sub&gt;r&lt;/sub&gt;′ [-]</td>
<td>1.37 0.96</td>
<td>1.11 0.76</td>
</tr>
<tr>
<td>Taylor micro-scale</td>
<td>λ&lt;sub&gt;y&lt;/sub&gt; [mm]</td>
<td>- -</td>
<td>0.066 -</td>
</tr>
<tr>
<td>Taylor-Reynolds number</td>
<td>Re&lt;sub&gt;λ&lt;/sub&gt; [-]</td>
<td>- -</td>
<td>83 -</td>
</tr>
</tbody>
</table>
As shown in Fig. 3.2, $\langle U_z \rangle$ profiles measured in the absence of a spray gradually increase from the fuel rod wall to a region with nearly constant velocity, hereafter referred to as 'coflow core'. For case A$_{II}$, the $\langle U_z \rangle$ profiles exhibit a different behavior in that $\langle U_z \rangle$ is significantly augmented close to the fuel rod wall. This increase is accompanied by a significant augmentation of $\langle U_r \rangle$ as well (see Table 3.3). This behavior is caused by the spray which accelerates the coflow in the near-wall region. The presence of the spray also induces a significant increase of the degree of anisotropy near the wall and a considerable decrease in the coflow core. Fig. 3.3 shows that $\langle U_z \rangle$ profiles for case H$_{II}$ exhibit a similar behavior, i.e. an acceleration of the coflow takes place in the wall region. The value of $u'_z/\langle U_z \rangle$ is significantly higher than in case A$_{II}$ in the wall region.

Figure 3.4: Mean oxygen volume fraction $\langle X_{O_2} \rangle$, mean temperature $\langle T \rangle$ and root-mean-square temperature fluctuations $T'$ profiles for case H$_{II}$ at $z = 0$ mm.
It is also observed for cases $A_{II}$ and $H_{II}$ that at the most outer parts of the coflow $u_z'/\langle U_z \rangle$ increases substantially. This evidence for a shear layer between the coflow and the laboratory air and it is of importance for case $H_{II}$. The laboratory air gradually mixes with the hot-diluted coflow changing the coflow inlet temperature and oxygen radial distribution. It will be shown in Section 3.3.5 that up to $z= 40$ mm, the coflow inlet profiles are preserved within the radial positions between $-40 < r < 40$ mm.

Information on the coflow mean temperature $\langle T \rangle$, r.m.s. temperature fluctuations $T'$ and oxygen volume fraction $\langle X_{O_2} \rangle$ is presented in Fig. 3.4. The $\langle X_{O_2} \rangle$ profiles in Fig. 3.4 show a symmetric and homogeneous coflow over a wide radial distance for case $H_{II}$. In the $\langle T \rangle$ profiles, $\langle T \rangle$ decreases towards the fuel rod wall due to the heat losses to the water-cooling system. In the near-wall region, asymmetries are seen on opposite side of the spray axis that exceed the inaccuracy of the CARS system (over 100 K at $r = 18$ mm). This is attributed to the different interaction of the premixed flames of the secondary burner close to the fuel rod wall. In addition, at the coflow annulus ($r = 80$ mm) a difference of 200 K is observed on opposite sides of the coflow. This results from a small offset of the vertical pipe relative to the fuel rod.

### 3.3.3 Liquid breakup mechanisms

High-speed visualizations of the liquid breakup were performed for cases $NA_{I}$, $A_{II}$ and $H_{II}$. Six sequential images are displayed in fig. 3.5 for case $NA_{I}$ and in fig. 3.6 for cases $A_{II}$ and $H_{II}$. Exposure time and camera frame rate were adjusted to fully resolve the liquid disintegration and to produce an optimal optical contrast. For case $NA_{I}$, the field-of-view has size $9.7 \times 3.0$ mm$^2$ and the frame rate was set to 40 kHz. The liquid breakup time scales for cases $A_{II}$ and $H_{II}$ differ from that of case $NA_{I}$. Therefore, the frame rate was increased to 120 kHz at the expense of reducing the field of view to $6.8 \times 3.0$ mm$^2$. The resolution of the imaging system was 26.6 $\mu$m per pixel for all the cases. The original video for each one of the three cases are available in the supplementary materials.

The Weber number for cases $NA_{I}$, $A_{II}$ and $H_{II}$ is 0.46, 3.26 and 0.46 respectively. For case $H_{II}$, a mean temperature gradient exists in the wall region, therefore, the thermophysical properties of the gas (needed to estimate the We number) were taken at $r = 11$ mm which are more representative of the conditions in the breakup region than those of the coflow bulk mean temperature.

Fig. 3.5a ($t=0s$) shows liquid emerging from the atomizer orifice in the form of a conical sheet. Liquid fuel passes through inclined tangential slots in the atomizer chamber inducing centrifugal forces that cause the liquid flow
3.3 Results and discussion

Figure 3.5: Sequence of images showing the liquid sheet breakup for case NA_1 (We = 0.46). The field-of-view has dimensions 9.68 x 3.04 mm².
to diverge radially. In the conical sheet, sinuous waves and short-wavelength surface disturbances are observed, each with different dynamics. Sinuous waves start near the orifice with low amplitude with respect to the wavelength. As the liquid sheet moves in the downstream direction (figs. 3.5a to 3.5d), the amplitude of the sinuous waves grows and the distance between successive short-wavelength disturbance crests increases. The sinuous wave amplitude growth leads to a thinning of the local sheet thickness between successive crests and small perforations of the liquid sheet occurs. As a rule, the liquid sheet disruption occurs in the regions with minimal sheet thickness yielding toroidal structures, lobes and small droplets.

The resulting fragments proceed into further breakup to a point where the restoring surface tension overcomes the disruptive aerodynamic forces and the breakup process ceases (see figs. 3.5e and 3.5f). Small perforations also occur in regions of the liquid sheet were no large wave amplitude are present. However, from the high-speed recordings, it is not clear whether such event results from the shear in the liquid-air interface or the liquid initial turbulence.

Similarly, for case $\text{A}_\text{II}$, small perforations occur in the conical sheet. These perforations rapidly evolve and yield toroidal structures that further disintegrate into droplets. However, the amplitude growth of sinuous waves at the liquid-air interface occurs faster than in the case $\text{NA}_1$ and the onset of breakup occurs closer to the atomizer orifice. This is due to the higher Weber number. The larger aerodynamic interactions between the liquid sheet and the surrounding gas, enables the growth of the surface waves and the eventual disintegration of the sheet.

The breakup mechanism for case $\text{H}_\text{II}$ differs from the previous cases. As the liquid emerges from the atomizer orifice (see figs. 3.6a and 3.6b), no wave amplitude growth is evident and a 'lump' of liquid immediately disrupts from the main liquid conical sheet. We believe that vaporization contributes to the local thinning of the conical liquid sheet. As the sheet thickness rapidly decreases, any perturbation causes the thin sheet to tear resulting in liquid lumps that subsequently disintegrate into ligaments and droplets (figs. 3.6c and 3.6e). It is also evident that some ligaments shrink and depending on their initial size and geometry may either form a single droplet or breakup into smaller lobes and 'satellite' droplets.
3.3 Results and discussion

3.3.4 Spray in Air Coflow Structure - Case $A_{II}$

Droplet Diameter distribution and Mean Velocity Field

Profiles of the $d_{32}$ at five axial distances, $z = 10, 20, 30, 40$ and $45$ mm along with the mean gas and droplet velocity fields are presented in Fig. 3.7. At each
axial station, radial transverses were performed in steps of 1 mm. The dashed line superimposed in the mean velocity field marks the locations where flame luminescence reaches a maximum. Hereafter, for case A\textsubscript{II}, the 'center-region' refers to the region near the axis surrounded by the adjacent inner flame-front whereas the 'outer-region' refers to the most outer part of the spray beyond the outer flame-front.

![Graph showing Sauter mean diameter Profiles and mean gas and droplet velocity fields for case A\textsubscript{II}. The dashed lines marks the maximum flame luminescence.](image)

At $z = 10$ mm, the maximum difference between $d_{32}$ values at corresponding points on opposite sides of the spray axis is 1.9 $\mu$m. Small droplets are present in the center-region and move mainly upwards. At larger distances from the spray axis, droplets size and average radial velocity increase indicat-
ing that a spreading of the spray into the coflow occurs. The data are consistent with the expected inertial response time associated with the droplet size. The small droplets resulting from the liquid jet breakup are responsive to the turbulence gas field and, therefore, are convected to the center due to the mean inward convective motion of the coflow caused by the spray presence. Above $z = 20$ mm an abrupt reduction of the $d_{32}$ is observed in the region between the inner and outer flame-front as a consequence of the rapid evaporation of the droplets caused by the appreciable heat-release from the oxidation process. As observed in the mean gas flow field, a significant increase of gas-phase velocity takes place in in this region as well. This supports the idea that oxidation occurs since the expansion of the combustion gases induces a local acceleration of the gas-phase. Large droplets are present at the outer-regions where no visible reactions are evident.

**Gas-phase velocity, temperature and droplet number density**

To study the influence of the droplets on the local turbulence structure and composition of the gas-phase, droplet number density $n_d$ along with the gas-phase mean axial velocity $\langle U_z \rangle_g$ and mean temperature $\langle T \rangle$ radial profiles are plotted in Fig. 3.8 at four axial stations, $z = 10$, 20, 30 and 45 mm. No CARS measurements were performed at axial stations below the lift-off height since the system inaccuracy exceeds the expected gas-phase temperature variation due to droplet evaporative cooling. To represent the large amount of data, the physical properties are jointly shown. The differently colored y-axis scales are used for the aforementioned physical quantities and the corresponding ranges have been preserved over the four axial stations. Droplets with diameter below 4 $\mu$m from the full record were taken as gas tracers for locations below the lift-off height. At the other axial stations the droplet diameter threshold was set to 8 $\mu$m. The results of the gas-phase are limited by the availability of the small droplets that fulfill the aforementioned constraints and each point presented in Fig. 3.8 is based on an ensemble of, at least, 100 samples. The error bars lying upon the dashed-dot line refer to the root-mean-square temperature fluctuations $T'$ and its scale is given at $z = 45$ mm, $r = 28$ mm. Fig. 3.9 displays the radial distribution of the root-mean-square temperature fluctuations $T'$ for three axial stations and complements the information given in Fig. 3.8.

At $z = 10$ mm droplets are mainly concentrated around $r = 6$ mm. The $\langle U_z \rangle_g$ profiles resemble a uniform, top-hat profile. The corresponding $\langle U_r \rangle_g$ radial profiles (not shown) are nearly null in the center-region indicating that the gas-phase moves predominantly in the axial direction.
Figure 3.8: Radial profiles of gas-phase mean axial velocity $\langle U_z \rangle_g$, mean temperature $\langle T \rangle$ and droplet number density $n_d$ at four axial stations for case $A_{II}$. The error bars represent the root-mean-square temperature fluctuations $T'$ and its respective scale is given at $z = 45$ mm, $r = 28$ mm.
From $z = 10$ mm to $z = 20$ mm, a decrease of $\langle U_z \rangle_g$ on the spray axis is accompanied by the spread of the $\langle U_z \rangle_g$ profiles as a consequence of the momentum flux from the spray center-region towards the periphery. Simultaneously, an increase of the droplet density occurs at larger radial distances from the axis. This results from the spreading of the droplets into the air coflow due to their initial droplet radial velocity component and relatively large Stokes number. CARS measurements were obtained even in the spray region with droplet density as high as $10^5$ cm$^{-3}$. A sharp increase of the local gas temperature and corresponding r.m.s. fluctuations (see Fig. 3.9) are observed in two regions indicating the presence of: 1) an inner flame-front ($9 < r < 11$ mm) that stabilizes inside the spray region and 2) an outer flame-front ($17 < r < 21$ mm) sustained by the air entrained and the turbulent mixing with the fuel vapor from droplets at the outer-region.

With increasing axial distance, a gradual decrease of $\langle U_z \rangle_g$ along the burner axis is observed. In the region delimited by the inner and outer flame-front, it is observed that along with a substantial decrease of the droplet number density initially present at $z = 20$ mm, $\langle T \rangle$ increases. This suggests that, as large droplets move across this region, fresh fuel vapor together reacts with the reminiscent oxygen in the mixture. Two observations can be made concerning the outer and inner flame-front: Firstly, the outer flame-front appears as a thin blue layer and moves farther away from the spray axis. This indicates
that a lean mixture results from the mixing of fuel vapor with the air coflow at increasingly larger radial distances due to the spreading of the spray droplets. Secondly, the inner flame-front moves closer to the spray axis and a reduction of \( n_d \) takes place. It is hypothesized that the inner flame-front motion towards the center-region is related to the mechanism of flame propagation between droplets and the velocity at which it occurs is much influenced by the effective fuel-air ratio, i.e., droplet-vapor-air ratio \([6, 7, 13, 38]\). Specifically, the combustion products formed in the region between the flame-fronts are transported by diffusion and convection towards the center-region. The heat of the combustion products increase the local rate of supply of the fuel vapor that eventually mixes with the surrounding gas. Reaction subsequently takes place and the inner flame-front propagates towards the spray axis. However, droplets acts as heat sink and depending on the local number of droplets, inter-droplet distance and turbulence characteristics, the amount of heat-release may not suffice to sustain the minimum temperature required to evaporate sufficient fuel vapor and, consequently, the flame propagation is halted.

**Temperature histograms**

![Temperature histograms](image)

Figure 3.10: Normalized temperature histograms across the spray outer-region at \( z = 30 \text{ mm} \) (\( \Sigma N = 1000 \text{ samples} \)) for case \( A_{II} \).
Normalized temperature histograms for five radial positions at \( z = 30 \) mm are presented in Fig. 3.10 for case AII. The total number of samples in each histogram amounts to 1000. A bin size of 50 K was selected for all the histograms presented. For each of the five radial positions selected, the corresponding mean \( \langle T \rangle \), root-mean-square temperature fluctuation \( T' \) as well as skewness \( S \) and kurtosis \( K \) of the temperature distribution are presented in the table located on the top-left. Negative values of skewness indicate data that are skewed towards lower temperatures and that the left tail is long relative to the right tail. The high kurtosis values indicate that the distribution has a sharper peak and longer, fatter tails.

In the outer flame-front, \( r = 22 \) mm, the temperature distribution is rather broad with temperatures extending from 2400 K down to 1400 K. A considerable amount of samples are over 2000 K. For radial positions between the flame-fronts, i.e. \( r = 20 \) mm and \( r = 16 \) mm, \( \langle T \rangle \) progressively decreases and the temperature distribution narrows significantly. At \( r = 12 \) mm a broadening of the observed temperature distribution along with a substantial number of samples over 2000 K is observed. In the inner flame-front, \( r = 9 \) mm, the number of peak temperatures increases as compared to \( r = 12 \) mm.

**Droplet properties per size class**

To illustrate the characteristics of the droplets behavior, the mean velocity \( \langle U_z \rangle \) of four droplet size-classes at three axial stations, \( z = 10, 20 \) and 45 mm, are shown in Fig. 3.11. The class width was chosen as 10 \( \mu \)m. For the axial station below the lift-off height, \( z = 10 \) mm, droplets with diameter below 4 \( \mu \)m are considered flow tracers and the mean velocity representative of the gas-phase. For the remain axial stations, a threshold diameter of 8 \( \mu \)m was selected. All the points are an average of at least 200 samples. The difference between the mean velocity for a particular size class and the gas phase mean velocity is called the slip velocity.

At \( z = 10 \) mm, it is observed that for larger droplets, the slip velocity relative to the gas-phase increases. This occurs as expected, since the Stokes number and, therefore, the respective inertial effects increase with the droplet diameter. Droplet \( \langle U_z \rangle \) profiles present a distinct behavior depending on the droplet diameter. Droplets smaller than 20 \( \mu \)m, display an almost monotonic decrease of \( \langle U_z \rangle \) with increasing radial distance from the spray center axis and the slip velocity is nearly zero in the center-region. For droplets larger than 30 \( \mu \)m, \( \langle U_z \rangle \) profiles show a single maximum at a radial position close to the center axis.

Three notable features can be observed on the droplet \( \langle U_z \rangle \) profiles with
increasing axial distance are as follows. First, in the center-region droplets smaller than 30 µm accommodate to the local gas-phase velocity. Secondly, droplets may 'survive' the region between both flame-fronts and eventually reach the air coflow, end up with nearly zero slip velocity and being carried upwards. Third, at the outer-region droplets smaller 30 µm tend to accelerate. The explanation is that due to the gas expansion and low initial droplet slip velocity, a reversal of the initial momentum flux, i.e. from the gas to the droplets, is established yielding an increase of those droplet size-class mean axial velocity.

Figure 3.11: Mean axial velocities $\langle U_z \rangle$ radial profiles per size class for case A_{II}. 

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Conventional and hot-diluted combustion regime
3.3 Results and discussion

3.3.5 Spray in Hot-diluted Coflow Structure - Case H\textsubscript{II}

The following section concerns the results of flame H\textsubscript{II} and they are presented in the same manner as flame A\textsubscript{II}. The most notable features of flame A\textsubscript{II} are recalled whenever appropriate so as to compare with the distinct peculiarities of flame H\textsubscript{II}.

**Droplet Diameter distribution and Mean Velocity Field**

Profiles of $d_{32}$ at five axial distances, $z = 10, 15, 20, 30$ and $40$ mm along with the mean gas and droplet velocity fields are presented in Fig. 3.12 for case H\textsubscript{II}.

Figure 3.12: Sauter mean diameter ($d_{32}$) profiles and mean gas and droplet velocity fields for case H\textsubscript{II}. The dashed lines marks the maximum flame luminescence.

Flame luminescence is marked by the dashed line superimposed on the mean velocity fields. As in case A\textsubscript{II}, an outer flame-front is visible and at the most outer parts of the spray, a region of droplets were no reaction is evident.
from the luminescence is observed. This region will be referred to as 'outer-region'. Droplets are virtually absent for locations above \( z = 45 \text{ mm} \) whereas in case HII droplets are still present for higher axial stations.

The \( d_{32} \) distribution at \( z = 10 \text{ mm} \) qualitatively resembles that of case AII. \( d_{32} \) is about 10 \( \mu \text{m} \) in the spray axis and increases for large radial distances from the spray axis. With increasing axial distance, droplets spread into the hot-diluted coflow and a substantial portion of spray becomes void of droplets in the center.

**Gas-phase velocity, temperature and droplet number density**

Fig. 3.13 shows the gas-phase mean axial velocity \( \langle U_z \rangle_g \) and the mean temperature \( \langle T \rangle \) along with droplet number density \( n_d \) at four axial stations for case HII. Due to the high droplet density in the region close to the spray nozzle, only a small number of valid CARS samples were acquired at \( z = 10 \text{ mm} \) and, therefore, no mean temperature profiles are presented for that axial station. Each of the different physical quantities is represented by the different colored y-axis scale and the scale is the same for the four axial stations. Fig. 3.14 displays the root-mean-square temperature fluctuations \( T' \) radial distribution at four axial stations and complements the information presented in Fig. 3.13.

In Fig. 3.13 it is observed that at \( z = 10 \text{ mm} \) droplets are mainly concentrated around \( r = 7 \text{ mm} \) and the \( n_d \) values are small as compared to those of case AII. \( \langle U_z \rangle_g \) profiles exhibit a 'dip' in the spray axis contrasting to what is observed in case AII (see Fig. 3.8).

At \( z = 15 \text{ mm} \), the presence of droplets is evident at a larger distances from the spray axis. This indicates that the droplet motion bears resemblance to that of case AII in that droplets spread into the coflow due to their initial outward radial momentum. A mean temperature difference of about 500 K is observed from the coflow core to a single peak located at \( z = 15 \text{ mm}, r = 13 \text{ mm} \). \( \langle T \rangle \) gradually decreases from the peak temperature towards the spray axis where the observed values are lower than that of the coflow, yet, significantly higher than the ethanol ignition temperature \( (T_{ig} = 653 \text{ K}) \).

It is hypothesized that local temperature in the spray region depends much on the initial spatial distribution of the droplets. As the liquid fuel breaks up into droplets, the hot-diluted coflow is entrained towards the spray axis. The entrained coflow initially encounters an increasing number of droplets and, therefore, available surface area for heat and mass transfer. A significant amount of energy is needed to overcome the liquid latent heat and vaporize the liquid droplets. Hence, the gas-phase temperature decreases from the coflow core to lower values at the spray axis. Likewise, the mixture fraction
Figure 3.13: Radial profiles of gas-phase mean axial velocity $\langle U_z \rangle_g$, mean temperature $\langle T \rangle$ and droplet number density $n_d$ at four axial stations for case $H_{II}$. The error bars represent the root-mean-square temperature fluctuation $T'$ and its respective scale is given at $z = 30$ mm, $r = 35$ mm.
increases towards the center of the spray. Since the high oxygen dilution in the oxidizer stream causes a shift of the peak adiabatic temperature towards leaner mixtures, a flammable mixture is promptly formed at the outer-region. Furthermore, the hot-diluted coflow consists mostly of nitrogen, which is basically inert as far as the bulk chemical heat-release is concerned, the combustion temperature and, hence, heat-release are reduced because of the expenditure of thermal energy to heat up nitrogen and other inerts. As a result, the temperature gradients observed at the outer flame-front are smaller than those of case A_{II}. Towards the spray axis, the amount of vapor diluted in the oxidizer becomes larger and the heat-release is gradually reduced yielding lower mean temperatures.

From $z = 15$ mm to $z = 30$ mm, a simultaneous process of droplet spreading and evaporation occurs yielding a progressive widening of the spray flame. This is evident from the observed evolution of the peak temperature location and the presence of droplets at larger distances from the center axis for more downstream axial stations. The combustion products formed at the outer flame-front are entrained towards the spray axis and dilute further the mixture near the spray axis.

Fig. 3.14 reveals three additional features of case H_{II}. First, at $z = 30$ mm,
3.3 Results and discussion

A peculiarity in the mean and r.m.s. temperature fluctuations radial profiles is observed. Between \( r = 0 \) and \( 8 \text{ mm} \), \( T' \) increases to a local peak and decreases to smaller values until \( r = 14 \text{ mm} \). At \( r = 8 \text{ mm} \), a substantially increase of \( \langle T \rangle \) is also observed (see Fig. 3.13). The radial profiles at \( z = 40 \text{ mm} \) are shown to prove that this observation is a real physical effect and not mere experimental error. This indicates that, as in case A\(_{II} \), an inner flame-front is also present. However, for case H\(_{II} \), as the spray evolves downstream the heat-release is significantly suppressed and no appreciable \( \langle T \rangle \) increase is observed at the inner flame-front location. Secondly, at \( z = 30 \text{ mm} \), in the outer flame-front region \( T' \) are only higher by 100 K over the temperature fluctuations of hot-diluted coflow core. However, \( T' \) remains about three times smaller than the values observed in the outer flame-front of case A\(_{II} \) at the same axial location (see Fig. 3.9). Last, at \( z = 30 \text{ mm} \), the region of the outer flame-front \( (14 < r < 27 \text{ mm}) \) is larger than the one of case A\(_{II} \) \( (22 < r < 27 \text{ mm}) \) indicating that the characteristic flame thickness differs in both cases.

Fig. 3.13 shows additional information concerning the influence of air entrainment on the coflow inlet temperature profiles. The laboratory air gradually mixes with the hot-diluted coflow changing the coflow initial radial temperature and oxygen distribution. It can be noticed that the coflow temperature at \( r = 40 \text{ mm} \) remains nearly the same throughout the axial stations presented. This shows that, for the range of radial distances displayed, the burner pipe diameter is wide enough to isolate the spray flame from the laboratory air entrainment. \[10!\] \( \{10!\} \) measurements at higher axial stations (not shown) indicate that above \( z = 60 \text{ mm} \) the outer region of the spray flame is perturbed by the laboratory air. Thus, below \( z = 60 \text{ mm} \), the spray flame is stabilized only by the hot-diluted coflow. Above the axial station of 60 mm, the oxidizer stream consists of a mixture of laboratory air together with hot-diluted coflow and reaction zone weakening effects may be present [81].

LDA measurements were carried out along the spray axis to better clarify the gas flow field. Gas-phase mean axial velocity \( \langle U_z \rangle_g \) and turbulent intensity \( u'^2_z/\langle U_z \rangle_g \) are presented in Fig. 3.15. A LDA system can measure the velocities of both \( \text{Al}_2\text{O}_3 \) seeding particles that faithfully follow the gas-phase and of larger droplets, hence, 'cross-talk' from the velocity distribution of the droplets to that of the seeding particles results in significant velocity bias in the continous phase statistics [101]. To avoid this, measurements were only carried out at axial positions above \( z = 20 \text{ mm} \), where no droplets are present.

An increase of \( \langle U_z \rangle_g \) can be observed along with a gradual decrease of \( u'^2_z/\langle U_z \rangle_g \). At \( z = 10 \text{ mm} \), \( \langle U_z \rangle_g \) profiles exhibit a 'dip' near the spray axis (see Fig. 3.13). These results show that the initial \( \langle U_z \rangle_g \) gradients lead to turbulent diffusion of momentum in the center, yielding an enhancement of
Figure 3.15: Gas-phase mean axial velocity $\langle U_z \rangle_g$ and turbulent intensity $u'_{zg}/\langle U_z \rangle_g$ evolution along the spray axis for case H_{II}.

the axial momentum along the spray axis.

**Temperature histograms**

The trend of the normalized temperature distribution from the coflow (r = 40 mm) towards the spray axis (r = 0 mm) at z = 30 mm are presented in Fig. 3.16. A bin size of 50 K was selected for all the histograms presented. Along with each histogram, the corresponding mean temperature $\langle T \rangle$ and r.m.s. temperature fluctuations $T'$ is given. Skewness (S) and kurtosis (K) of the temperature distribution are presented in the legend on the top-left side.

At r = 40 mm, $\langle T \rangle$ and $T'$ are 1421 K and 105 K, respectively. At the outer flame-front, r = 21 mm, $T'$ is only about 50 K higher than that of the coflow and the normalized temperature histograms shape are rather alike, yet, shifted. It is also observed that the number of samples above $T = 2000$ K is reduced as compared to the ones observed in case A_{II} (see Fig. 3.10). For r = 10, 8 and 0 mm, $\langle T \rangle$ successively decreases and only minor changes on the normalized histograms are observed. It is evident that in the region of the inner flame-front, r = 8 and 10 mm, no peak temperatures above 2000 K are present. At r = 0 mm a substantial reduction of $T'$ is observed and the temperature distribution is symmetric (S = 0). Temperature histograms along
3.3 Results and discussion

Figure 3.16: Normalized temperature histograms for five radial positions at $z = 30$ mm ($\Sigma N = 1000$ samples).

the spray axis above $z = 20$ mm (not shown) have virtually identical skewness value. This indicates that, as a result of the turbulent mixing, the gas-phase is rather homogeneous in the region of the spray axis.

**Droplet properties per size class**

Velocity profiles of selected size classes of droplet ensembles are presented in Fig. 3.17 for case $H_{II}$. The development of $\langle U_z \rangle$ for case $H_{II}$ shows that, similar trends at $z = 10$ mm hold, i.e. increasing droplet slip velocity with the size-class is observed. An higher slip velocity of droplets between 10 and 20 \( \mu \)m is observed. Farther downstream, slip velocity gradually decreases and at $z = 30$ mm the different size-class radial profiles exhibit a similar mean axial velocity.
3.4 Spray phenomenology in air and hot-diluted coflow

Fig. 3.18 presents a schematic of the spray structure of \( A_{II} \) and \( H_{II} \) cases. The goal of this section is to provide a summary of the most notable features of both cases and elaborate a schematic illustration of the spray structure of both cases.

Fig. 3.18 presents a schematic of the spray structure of \( 12! \) and \( 13! \) cases. The goal of this section is to provide a summary of the most notable features of both cases and elaborate a schematic illustration of the spray structure of
3.4 Spray phenomenology in air and hot-diluted coflow

Figure 3.18: Schematic of spray structure of pressure-swirl sprays in air and hot-diluted coflow. The dash-dotted line represents the gas-phase mean temperature radial profile along the spray cross section.

In the air coflow case, Fig. 3.18a, the presence of the spray induces entrainment of the air coflow into the spray. As a result of the liquid breakup and the inward convective motion of the coflow generated by the spray, small droplets are mainly located in the center whereas large droplets are encountered at the most outer parts of the spray. While the large droplets spread into the air coflow and small droplets move upwards, the difference in velocity between gas and droplet enhances the droplets vaporization rate and fuel vapor is produced. An inner and an outer lean flame-front develops. The outer flame-front is sustained by the entrainment of air coflow and the fuel vapor from the large droplets at the most outer parts of the spray. The inner flame-front appears in a region inside the spray and gradually moves towards the center-region wherein a mixture of air and droplets and vapor fuel is present. It is hypothesized that this takes place through flame propagation mechanics and the speed at which it occurs depends on the local droplet-vapor-air mixture and gas turbulent characteristics. Furthermore, in the region between both flame-
Conventional and hot-diluted combustion regime

fronts, the fresh fuel vapor from the existing droplets and remaining oxygen in the combustion products may react further. Throughout most of the spray, a substantial number of temperature samples above 2000 K is observed.

In the hot-diluted coflow case, Fig. 3.18b, the initial droplet $d_{32}$ distribution resembles in many aspects that of the air coflow. The differences observed in the gas-phase velocity and temperature fields are due to the presence of strong vaporization rates which, consequently, yield a distinct turbulence modulation of the gaseous phase in the near atomizer region. As a result of the entrainment of hot-diluted coflow, droplets are embedded in a hot oxygen reduced gaseous phase and evaporate at the cost of heat losses from the gaseous phase. Hence, the temperature decreases towards the spray center. The temperature decrease is accompanied by an increase of fuel vapor towards the spray axis yielding a premixed region at the center. The significant oxygen dilution in the gaseous phase causes a shift of the stoichiometric mixture fraction towards leaner mixtures. Thus, the mixture formed at the most outer parts of the spray yields a broad outer flame-front with a single temperature maximum. For the most inner parts of the spray, the mixture becomes increasingly rich yielding, therefore, progressively lower temperatures. Hence, a substantial reduction of temperature samples above 2000K is observed and the temperature rise of the inner flame-front is substantially smaller as compared to the air coflow case.

3.5 Conclusion

A comprehensive study was performed of combusting ethanol sprays with similar injection pressure in air and hot-diluted coflow conditions. The objective was to describe the features of spray combustion in conventional and hot-diluted combustion regimes. The experimental investigations involved high-speed visualization of the liquid jet breakup and complementary pointwise measurement techniques in the spray region. Measurements of droplet size and velocity components in the spray region were obtained by PDA. CARS measurements were carried out in the spray region. The data processing procedure used to take into account the effects of the presence of droplets on the CARS spectra provides reliable and consistent gas-phase temperature measurements.

The experimental results show that the presence of strong vaporization rates produces significant changes of liquid breakup mechanics and of the gas-phase mean velocity in the near atomizer region. The coflow temperature and oxygen dilution have a profound impact on the spray flame structure and, ultimately, on the temperature peaks observed. In air coflow, droplets are
embedded in air as gaseous phase. An inner and outer flame-front is observed. The inner flame-front propagates towards the small droplets embedded in an upward flow of air depending on the local vapor fuel-droplet distribution and flow conditions. In the hot-diluted coflow, the existence of a hot and diluted surrounding gaseous phase around the droplets instead of air has two effects: 1) a shift of the peak adiabatic temperature towards leaner mixtures and 2) enhancement of the droplets vaporization rates. The mixing of the droplet spray and the hot-diluted coflow, and the rapid evaporation together provide a radial gas composition profile with the gaseous mixture increasingly rich towards the spray center-region. This yields progressively lower temperatures toward the spray axis and a substantial reduction of peak temperatures.

The experiments reported here provide a valuable dataset for model validation and new opportunities compared to other datasets reported previously in the literature, as reviewed in Jenny et al. [73]. Because the experiments were done using a pressure-swirl atomizer instead of a nebuliser, they show the complexities of liquid desintegration and the transition from the dense to the dilute spray region, relevant in practical applications. The inlet boundary conditions for model simulations in the dilute region can be constructed from the droplet statistics from PDA as well as the gas-phase velocity statistics from LDA. The gas-phase temperature statistics are provided by CARS. Not available from measurements and hence to be supplied based on modeling assumptions are data on droplet temperature and vapor concentration. On other hand, the information concerning the liquid fuel pressure and temperature upstream the pressure-swirl atomizer together with the high-speed visualizations, provides a promising tool for fuel spray breakup models. The current work provides the data needed for a comparative modeling study of the sprays in air and in hot-diluted coflow.
CHAPTER 4

Effects of coflow conditions on ethanol spray flames in hot diluted coflow with varying degree of temperature and oxygen concentration*

Ethanol pressure-swirl sprays issuing in a hot-diluted oxidizer coflow with different temperature and composition were studied. The bulk coflow temperature was varied together with the oxygen volume fraction. The bulk coflow temperature was changed from 1480 K to 1225 K and the oxygen volume fraction from 7.1% to 10.1%. The liquid mass flow rates were chosen to yield spray flames with nearly identical Weber number. Laser Doppler anemometry, phase Doppler anemometry and coherent anti-Stokes Raman scattering were applied in the spray region and the coflow inlet. The current measurements provide a thorough description of the spray structure, droplet dispersion and gas temperature fields as well as a comprehensive database useful for validation of numerical models. Visual observations of the flame luminescence reveal that the lift-off height depends on the liquid mass flow rates as well as the coflow conditions. The lift-off height is shown to increase for lower coflow temperatures and higher liquid mass flow rates. It is found that lift-off behavior depends on the droplet convective, vaporization and chemical time scales prior to ignition. Phase Doppler anemometry results indicate that the droplet mean size and velocity distribution close to the atomizer are not influenced by the coflow conditions. A flame-front develops at the outer region of the spray where a low density of large droplets are present. A significant number of peak temperatures samples above 2000 K is observed at this location. Decrease of the coflow temperature leads to a reduction of the local peak temperatures. Closer to the center axis, the local mixture composition becomes increasingly rich and the heat-release is lower than in the outer region.

*The results of this chapter have partially been published in:
4.1 Introduction

MILD combustion has prompted great interest as a new clean combustion concept. The operation principle is based on the dilution of fuel with preheated air and high temperature combustion gas recirculation yielding an overall small temperature increase due to the heat capacity augmentation and, consequently, low NO$_x$ emissions [3, 49]. This unique combustion mode has completely different characteristics from a conventional operating method concerning flammability limits, combustion stability, combustion noise, and ignition process and has been studied both in industrial furnaces and laboratory scale setups for various gaseous fuels. Issues of interest for the studies included flame chemistry [61, 79, 80], flow dynamics [19, 31, 32], stabilization mechanisms [30], emissions formation and quantification of the thermal field uniformity under various operational conditions [15, 16, 76].

A review of earlier research show that MILD combustion for liquids fuels has received little attention [41, 60, 85, 91]. Studies have been performed mainly in industrial and semi-industrial scale furnaces and focused on the sustainability of MILD regime for heavy fuel oils as well as the factors resulting in furnace performance deterioration. Tsuji [41] investigated the applicability of heavy fuel oils in industrial furnaces operating in MILD conditions. The oxygen content and the pre-heated air temperature were varied systematically ($Y_{O_2} = 3 - 15\%$ and $T = 573 - 1553\, \text{K}$) to investigate the changing state of the flame form and the resulting pollutant emission. A strong dependence of the flame stability and chemiluminescence with the preheated air and oxygen dilution was observed as well as a substantial increase of NO$_x$ production for heavy fuel oils. Other studies [85, 91] in industrial scale test furnaces operating in MILD conditions showed a homogenized temperature field in furnaces. However, only for certain nozzle configurations and fuel oil grades a high-temperature process without the penalty of increased NO$_x$ emissions was achievable. Derudi [60] carried out an experimental study on a dual-nozzle laboratory scale burner arranged in a crossflow configuration. Pure liquid $n$-octane and other mixtures involving hydrocarbons commonly found in practical fuels were used. The experiments were designed to map the dependence of the MILD combustion region with two main operating parameters: average combustion chamber temperature and dilution ratio. Results on NO$_x$ and CO emissions showed that MILD conditions of liquid hydrocarbons was attainable for lower combustion gas recirculation ratio as compared to the LPG fuel counterparts. Additionally, increase of the oxygen dilution was witnessed to have a slight positive effect on pollutant emissions for lower
average combustion chamber temperature.

The aforementioned studies clearly show the practical importance of the technology, however, many aspects in the near-injector region are not investigated. In the spray region, the fuel stream first has to be shattered into an ensemble of droplets with a desirable distribution of sizes and velocities in order to achieve the required rate of vaporization, chemical heat-release, levels of conversion and pollutant emissions. Instabilities at the liquid-gas interface in the near-atomizer region lead to primary and secondary break-up. The liquid fragments in surrounding gas form a dispersed multiphase flow, where droplets are evaporating and subject to turbulent dispersion. The dispersed droplets modify the gas phase turbulence and the released fuel vapor depending on the relative speed of evaporation, mixing and combustion, burns in diffusion flames around individual droplets or clusters of droplets, or in partially premixed gaseous flames [73]. The temperature and composition of the oxidizer influence the time scales of phase change and reaction, making a spray flame in conventional combustion different from that of a hot-diluted combustion regime.

The present work concerns a fundamental experimental study. The objective is to reach understanding of the spray region by using a laboratory scale burner developed to study spray combustion in MILD conditions. The burner design retains the relevant physical processes of practical combustion systems. However, the composition of the gases entrained into the spray are controlled by a secondary burner rather than the furnace aerodynamics. The spray flame is unconfined and specifically designed to allow for easy access of state-of-the-art laser diagnostics [89].

Goix et al. [77], Cessou and Stepowski [4] and Stepowski [27] investigated stabilization of flame structures above an airblast injector fed with liquid methanol. The qualitative structure of the reacting spray and its reaction zone were investigated by Mie scattering imaging and OH* Planar LIF. Phase Doppler anemometry was used to measure droplet size statistics. The results show two outer reaction zones where strong OH signals are present. It is observed that flame stabilization occurs in the large scale mixing structures at the edge of the jet where the small droplets have sufficient time to vaporize, mix with entrained air, and burn. A discussion was presented concerning the lift-off position, motivated by a mixture-fraction formulation. In an effort to establish general features of combusting sprays in hot-diluted coflow without the influence of the near-injector dense region, O’Loughlin and Masri [99] used a nebuliser instead of a pressure atomizer. Simultaneous high-speed OH-CH* Planar LIF and droplet Mie scattering show that ignition OH kernels formation and growth is the mechanism of spray flame stabilization in hot-diluted
conditions. The oxygen mole fraction in the coflow (12% by volume) of \[99\] is, however, rather high compared to other studies of combustion systems operating in MILD conditions \[41, 60\].

This chapter is an extension of earlier work by the authors on the structure of pressure-swirl spray flames in conventional and hot-diluted combustion regime \[44\]. The burner facility, atomizer type and the liquid fuel used in this study were identical. A commercial pressure-swirl atomizer was used for generating the spray. In this way the liquid atomization and gas flow in the combustion chamber are very strongly coupled and experiments remain close to the most common configuration used in industrial practice. All measurements are made up to locations as close to the atomizer as possible to unveil features of the near-injector region. Ethanol was used as fuel because of the well-defined physical properties and well-documented chemical mechanism \[70, 72\].

In the previous chapter, measurements of profiles of droplet size and velocity and gas-phase temperature were performed in an axisymmetric spray issuing in air and in hot-diluted coflow. The results show that in a reacting spray in air coflow, an inner and an outer flame-front are present. In hot-diluted coflow, weakening of the inner flame-front occurs due to the local gaseous mixture richness. At the location of the outer flame-front temperature samples above 2000 K persist. In this chapter, three spray issuing in hot-diluted coflows with different temperature and composition were studied. The present study complements the previous chapter \[44\] and contributes to the manuscript by: (a) describing changes in the flame structure and identifying the driving parameter for peak temperature reduction in the outer flame-front region, (b) providing a comprehensive set of measurements that extend the initial database for model validation of polydispersed sprays, (c) provide insight into the physical mechanisms controlling the spray flame lift-off height in hot-diluted conditions.

This chapter is structured as follows: Section 4.2 presents the input parameters as well as the coflow boundary conditions for three test cases. Section 4.3 presents the results on spray flames visual appearance, lift-off behavior, droplet distribution and spray flame structure. A summary of the spray phenomenology and a discussion of the spray flame stabilization mechanisms is also given. In Section 4.4, conclusions are presented.

4.2 Cases description
Table 4.1: Burner operating conditions and dimensionless numbers of the flames studied. The pressure fluctuations are less than 0.2% for all flames.

<table>
<thead>
<tr>
<th>Case</th>
<th>Coflow</th>
<th>Spray</th>
<th>$\xi_{st}$ [-]</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>$\dot{m}_{\text{air}}$ [kg/h]</td>
<td>$\dot{m}_{\text{DNG}}$ [kg/h]</td>
<td>$U_{\text{cf}}$ [m/s]</td>
</tr>
<tr>
<td>$H_I$</td>
<td>70</td>
<td>3.28</td>
<td>3.42</td>
</tr>
<tr>
<td>$H_I^{\text{Aux}}$</td>
<td>1.53</td>
<td>15.9</td>
<td>304</td>
</tr>
<tr>
<td>$H_{II}$</td>
<td>51</td>
<td>2.13</td>
<td>2.23</td>
</tr>
<tr>
<td>$H_{II}^{\text{Aux}}$</td>
<td>1.63</td>
<td>14.6</td>
<td>301</td>
</tr>
<tr>
<td>$H_{III}$</td>
<td>45</td>
<td>1.75</td>
<td>1.81</td>
</tr>
</tbody>
</table>
Table 4.2: Summary of measurements locations for cases H_I, H_{II} and H_{III}.

<table>
<thead>
<tr>
<th>Measurement technique:</th>
<th>z (mm)</th>
<th>Comment:</th>
</tr>
</thead>
<tbody>
<tr>
<td>LDA</td>
<td>0, 20, 25, 30, 35,</td>
<td>Centerline,</td>
</tr>
<tr>
<td></td>
<td>40, 45, 50</td>
<td>Coflow radial profiles</td>
</tr>
<tr>
<td>CARS</td>
<td>0, 15, 20, 30, 40,</td>
<td>Coflow radial profiles</td>
</tr>
<tr>
<td></td>
<td>50, 60</td>
<td>Coflow radial profiles, Spray</td>
</tr>
<tr>
<td></td>
<td></td>
<td>region</td>
</tr>
<tr>
<td>Flue gas analyser</td>
<td>0</td>
<td>Coflow radial profiles</td>
</tr>
<tr>
<td>PDA</td>
<td>8, 10, 12, 15, 20,</td>
<td>Spray region,</td>
</tr>
<tr>
<td></td>
<td>30, 35, 40, 45</td>
<td>Steps of 1 mm</td>
</tr>
</tbody>
</table>

The operating conditions as well as relevant dimensionless numbers of each individual flame studied are given in Table 4.1. The positions where the measurement techniques were employed are summarized in Table 4.2. Three spray flames in different coflow conditions were thoroughly studied. The three flames are denoted H_I, H_{II} and H_{III}. In addition, some results are reported for two auxiliary cases H_{I_{Aux}} and H_{II_{Aux}} corresponding to sprays with higher liquid mass flow rates in identical hot-coflow conditions as H_I and H_{II}, respectively. These are included to elucidate the droplet velocity statistics in the near atomizer region and help the discussion of the flame lift-off behavior presented in Section 2?. Only PDA measurements were made for these two cases. \( U_{cf}, T_{cf} \) and \( \langle X_{O_2} \rangle \) represent the mass average velocity, temperature and oxygen volume fraction of the coflow, respectively. The values were determined by the measured velocity, temperature and oxygen volume fraction profiles at \( z = 0 \) mm, i.e. at the atomizer orifice exit plane. The Reynolds number of the coflow (\( Re_{cf} \)) was based on the hydraulic diameter \( (D_h = 158 \) mm) and the gas properties estimated from the bulk temperature and composition. The Taylor microscale \( \lambda_g \) was estimated at \( r = 15, 17 \) and \( 12 \) mm for case H_I, H_{II} and H_{III}, respectively. It was determined from autocorrelation functions constructed from time series of the axial velocity component with \( 2.5 \times 10^5 \) samples by using the slotting method with local normalisation [67]. Taylor's hypothesis was used to convert timescales into length scales.
The Weber number is defined as $\rho_g U_{jet}^2 l/\sigma$ and is identical for the three spray flames studied. $l$ is the characteristic liquid film thickness and it was assumed to be equal to the nozzle diameter, i.e. 210 µm. $U_{jet}$ represents the difference between the estimated liquid jet velocity from the liquid mass flow rate and $U_{cf}$.

The coflow velocity profiles at $z = 0$ mm are displayed on Fig. 4.1 to provide the input boundary conditions for numerical simulations [17] as well as relevant information to understand the spray flame behavior. On the left-hand side, the coflow axial mean velocity $\langle U_z \rangle$ and the respective r.m.s. fluctuations $u'_z$ are presented. The same quantities are shown for the radial velocity component on the right-hand side. It is observed that, on opposite sides of the spray, for all cases the magnitude of $\langle U_r \rangle$ sharply increases until a maxima in the near-wall of the central rod and then gradually decreases outwards. This indicates that the hot-diluted coflow has a strong inward velocity component and is being convected towards the spray axis filling in the void space between the ligaments and droplets formed at the center.

The r.m.s. fluctuations profiles of the axial and radial velocity components, $u'_z$ and $u'_r$, show a sharp increase between $r = 70$ and 80 mm. This indicates that a shear layer is formed between the hot-diluted coflow and the laboratory air. Thus, the gradual mixing of laboratory air with the hot-diluted coflow

![Figure 4.1: Coflow mean velocities and r.m.s. fluctuations profiles of the axial and radial velocity components at z = 0 mm. (—— : H_I , —— : H_{II}, ——— : H_{III})](image-url)
causes changes in the radial profiles of temperature and oxygen concentration and, ultimately, perturbs the spray flame. It will be shown in Section 4.3.6 that up to \( z = 60 \text{ mm} \), the burner pipe diameter is wide enough to isolate the spray flame from the laboratory air entrainment.

Fig. 4.2 displays the coflow mean temperature \( \langle T \rangle \) and r.m.s. fluctuations \( T' \) as well as the oxygen volume fraction \( \langle X_{O_2} \rangle \) for the three cases presented in Table 4.1. For the cases studied, \( \langle X_{O_2} \rangle \) radial profiles are fairly uniform over a significant radial distance in the coflow. In the temperature profiles, asymmetries are seen that exceed the inaccuracy of the CARS system. The maximum difference between the \( \langle T \rangle_{max} \) at opposite sides of the spray is observed in case \( H_{II} \) and amounts to 34 K.

![Figure 4.2: Mean oxygen volume fraction \( \langle X_{O_2} \rangle \), mean temperature \( \langle T \rangle \) and root-mean-square temperature fluctuations \( T' \) profiles at \( z = 0 \text{ mm} \). (--- : \( H_I \), — : \( H_{II} \), —— : \( H_{III} \))](image-url)
4.3 Results and discussion

4.3.1 Spray flames visual appearance and behavior

To show the overall flame appearance, color pictures are displayed in Fig. 4.3. The flames were photographed using a digital camera (Canon EOS X5) and have 0.6 s exposure time and identical aperture. The right inset shows a close-up of the spray region and initial development of the flame. For all the flames presented, the field-of-view of the inset measures 170×160 mm² (width×height).

It is observed that the atomizer yields conical spray flames. Based on the visual appearance, the spray flames in hot-diluted coflow are comprised of three regions: jet liquid break-up with no visible flame luminescence, a bluish reaction zone and a yellow zone farther downstream. The images shows that flame H_{III} has a stronger luminescence. Furthermore, a shorter flame length is observed as compared to flames H_I and H_{II}. For all the cases, the insets show a region of bright luminescence in the outer regions of the spray.

![Fire images](image_url)

Figure 4.3: Images of spray flames studied with 0.6 s exposure time and identical aperture. The bulk properties of the coflow inlet for each spray flame are listed below.

- Flame $H_I$:
  - $U_{ef} = 3.42$ m/s
  - $T_{ef} = 1480K$
  - $\langle X_{O_2} \rangle = 7.1\%$

- Flame $H_{II}$:
  - $U_{ef} = 2.24$ m/s
  - $T_{ef} = 1300$ K
  - $\langle X_{O_2} \rangle = 9.3\%$

- Flame $H_{III}$:
  - $U_{ef} = 1.81$ m/s
  - $T_{ef} = 1225$ K
  - $\langle X_{O_2} \rangle = 10.1\%$
A study was performed to determine the lift-off height dependence on the liquid mass flow rates for the coflow conditions of case $H_I$, $H_{II}$ and $H_{III}$. Experiments were also conducted for spray flames in air coflow and have been reported in [44]. In the current study, the procedure to determine the lift-off height was based on average flame luminescence from the photographs. It is defined as the axial distance from the atomizer orifice exit plane, $z = 0$ mm, to the lowest axial position of the bluish reaction zone along the spray axis.

The outcome is presented in Fig. 4.4 and the coflow conditions are presented in the inset. The test cases of this study are identified in the figure by the dashed squares. Case $A_{II}$, a reacting spray in air coflow, was previously described by the authors [44] and is also presented. When the liquid mass flow rates were below 1.10 kg/h, pre-vaporization of the ethanol occurred yielding a yellowish flame anchored at the atomizer orifice exit plane. Liquid mass flow rates higher than 1.70 kg/h could not be achieved with the current experimental setup.

As shown in Fig. 4.4, the lift-off height of sprays in hot-diluted conditions depends linearly on the liquid mass flow rate whereas in air coflow a non-linear dependence is observed. This is evidence for the existence of different
stabilization mechanics of the spray flames in hot-diluted coflow. Furthermore, it is observed that in the coflow conditions of case H_{III}, the lift-off height is more sensitive to the liquid mass flow rate than in the conditions of cases H_{I} and H_{II}.

Secondary breakup is not expected to play an important role in these flames. For the sprays in air coflow the Weber number varies from 2 to 3.4 and does not exceed a critical Weber number of 12 (based on [8]). Thus, in air coflow the secondary breakup is expected to be absent. In hot-diluted conditions, Weber numbers are smaller and within the range of liquid mass flow rates studied only vary from 0.2 to 0.5. Although, to our knowledge in the literature no information is available on the secondary breakup modes in the presence of strong evaporation rates, the ratio of the aerodynamic forces and surface tension remains significantly below unity and, therefore, no changes on the secondary breakup regime are expected.

### 4.3.2 Droplet size and mean velocity field

Fig. 4.5 shows droplet Sauter mean diameter \(d_{32}\) and mean axial \(\langle U_z \rangle\) and radial \(\langle U_r \rangle\) velocity profiles for cases H_{I}, H_{II} and H_{III} at several axial stations. For flame H_{I}, no droplets are observed above the axial station of 30 mm. Regions in the spray core where no data are plotted correspond to positions where acquisition of sufficient amount of data within a reasonable measuring time was not possible.

The experimental results indicate that the initial \(d_{32}\) and velocity initial profiles as well as subsequent evolution downstream are similar for the three flames studied. At \(z = 10\) mm the sprays are non-uniform with smaller \(d_{32}\) near the centerline and large \(d_{32}\) at the periphery. It is observed that droplets have a zero \(\langle U_r \rangle\) close to the centerline. From the centerline until the spray periphery, \(\langle U_r \rangle\) magnitude gradually increases until a maximum. This indicates that droplets are spreading into the hot-diluted coflow and it is in agreement with the visual appearance of the spray flames (see Fig. 4.3 insets). With increasing radial distance, \(\langle U_z \rangle\) increases until a maximum and then decreases to nearly the same magnitude as in the center region. For all the cases, a good symmetry of both velocity components and \(d_{32}\) is observed on opposite sides of the sprays axis.

With increasing axial distance from the atomizer, the center region of the sprays become void of droplets. As expected, this effect is more pronounced for flame H_{I} since \(T_{cd}\) is the highest and droplets evaporate faster. A gradual reduction of the droplet velocity components is observed due to the momentum exchange of the droplets with the gaseous phase. An increase in
the $d_{32}$ between $z = 10$ mm and $z = 30$ mm is observed. The reason for this is two-fold. First, the quicker vaporization of the small droplets fraction in the droplet ensemble reduces significantly the surface area. Secondly, small drag/inertial effects of large droplets leads to the displacement of the droplets outwards, progressively increasing the liquid fuel mass available in the spray peripheral regions of the spray.

### 4.3.3 Spray flames initial droplet distribution

To demonstrate the importance of the initial conditions on the overall spray morphology and development, the results of polydisperse ensemble of drops at $z = 10$ mm is plotted in Fig. 4.6. The three radial positions selected correspond to the spray centerline, the radial position where the droplet number concentration is maximum (see Fig. 4.7) and the spray edge. For case $H_1$, no droplets are present at $r = 0$ mm and the radial position of 2 mm was se-
lected as representative of the centerline. Droplet size frequency distribution curves (left y-axis) are overlapped with the corresponding cumulative droplet distribution curves (right y-axis). The distribution curves based on number of droplets per size class are represented by black lines whereas gray lines refer to volume size and can be related to mass by the liquid fuel density. Note that the droplet distribution based on volume are skewed to the right as expected due to the weighting effect of the larger droplets.

The results show clear differences between the droplet distributions at the radial positions presented, however, the trend among the three cases studied are in common. Flame $H_I$ shows that, at $r = 2$ mm, 50% of the droplets are below 5 $\mu$m indicating that a high droplet surface area is available. 50% of the droplet mass is contained in the fraction below 18 $\mu$m. At larger radial distances from the spray centerline, $r = 6$ mm and $r = 13$ mm, the spread of the droplet number distribution increases and more larger droplets are observed.

Figure 4.6: Frequency distribution curves (solid lines) and cumulative droplet size distribution (dashed lines) based on number ($d$) and volume ($d^3$) at $z = 10$ mm for flames $H_I$, $H_{II}$ and $H_{III}$.
The droplet diameter below which 50% of the mass is contained also increases.

Regarding flames $H_{II}$ and $H_{III}$, it is observed that at all the three radial positions larger droplets are more abundant in the droplet size distributions than in $H_{I}$. At the centerline, $r = 0$ mm, 50% of the droplet mass resides in droplets below 28 $\mu$m and 24 $\mu$m for flame $H_{II}$ and $H_{III}$, respectively. At $r = 6$ mm it can be noticed that, as compared to flame $H_{I}$, droplet number cumulative distributions do not change significantly. For the radial position of $r = 13$ mm and 14 mm a considerable number of large droplets is clearly observed and, therefore, more fuel mass is available at the spray periphery for cases $H_{II}$ and $H_{III}$.

### 4.3.4 Droplet number density

The collection of liquid droplets spreads through the gaseous environment according to their initial momentum and droplet-gas interactions. Radial profiles of the droplet number density ($n_d$) at several axial stations are shown in Fig. 4.7 in order to elucidate the degree of dispersion and identify regions where gas-phase composition and momentum may be significantly influenced by the droplets presence.

![Figure 4.7: Variation of droplet number density $n_d$ with the radial distance measured at different axial positions for flames $H_{I}$, $H_{II}$ and $H_{III}$.](image-url)
For all cases, at z = 10 mm, \( n_d \) is nonuniform across the radial traverse and droplets mainly are concentrated at \( r = 6 \) mm. The subsequent development shows a broadening of the spray with increasing axial distance and a significant reduction of the droplet number density. Furthermore, in the region close to the spray axis no droplets are present. This effect is more significant for case H1. In all the cases studied, the largest radial distance from the spray axis at which droplets are observed is nearly the same.

4.3.5 Gas-phase flow field

The evolution of the gas-phase mean axial \( \langle U_z \rangle_g \) and radial \( \langle U_r \rangle_g \) velocity components and normal \( \langle u_z u_z \rangle_g \) and shear \( \langle u_z u_r \rangle_g \) Reynolds stress profiles [56] between four axial stations is displayed in Fig. 4.8. The subscript "g" denotes for the gas-phase. Each point presented in Fig. 4.8 is based on an ensemble of, at least, 100 samples. This threshold was found by investigating the convergence of the velocity mean and higher order statistical moments of droplets smaller than 6 \( \mu \)m as the number of samples increases. Notice that profiles of the physical quantities presented are similar for the different cases and the curves have been shifted for clarity. Due to the striking similarity among the profiles the following observations are common for all the three cases studied.

The \( \langle U_z \rangle_g \) profiles indicate that at \( z = 10 \) mm a peak value occurs at about \( r = 6 \) mm. In the region around the centerline, \( \langle U_z \rangle_g \) magnitude is close to the small droplets axial velocity (see Fig. 4.5) indicating that droplets move along with the gas flow field and no substantial momentum exchange through drag takes place in the center. At downstream axial stations, the \( \langle U_z \rangle_g \) magnitude gradually decreases across the spray domain. This occurs as a result of the turbulent mixing with the entrained hot-diluted coflow. Moreover, the number droplet density is relatively high at this radial position. Hence, a modulation of the gas velocity field by the droplets is expected.

The \( \langle U_r \rangle_g \) magnitude is one order of magnitude smaller than \( \langle U_z \rangle_g \) at \( z = 10 \) mm. Further downstream, a reduction of the radial velocity component takes place to a nearly nil value. This shows that the gas flow field move mainly upwards and droplets shoot outwards. Depending on their initial size diameter and momentum, the droplet trajectory path is deflected towards the direction parallel to the spray axis.

Both Reynolds-stresses, \( \langle u_z u_z \rangle_g \) and \( \langle u_z u_r \rangle_g \), are high close to the exit plane of the atomizer. Moreover, a large difference between \( \langle u_z u_z \rangle_g \) and \( \langle u_z u_r \rangle_g \) components is observed, with \( \langle u_z u_z \rangle_g \) substantially higher than \( \langle u_z u_r \rangle_g \). That can be attributed to the effect of liquid break-up in the dense region. Ad-
Additionally, it indicates that significant mixing takes place in the near atomizer region. A fast decay of fluctuation levels until 30 mm is observed resulting in small anisotropy in the regions farther downstream.

Since the number of droplets is very low in the center region of the spray, LDA measurements were conducted along the spray centerline to determine the gas-phase velocity. The LDA system can measure the velocities of both 'seeding' particles that faithfully follow the gas-phase and of droplets. Therefore, 'cross-talk' from the velocity distribution of the droplets to that of the particles potentially results in velocity bias in the gas-phase statistics [101].
The degree of bias will depend on the correlation between the particle and droplet arrival rate. In view of that, measurements were performed only in positions were no droplets are present, i.e. in the center region void of droplets. Additionally, prior to any further estimation of the flow velocity component, velocity distribution were meticulously studied to look for the evidence of outliers.

The evolution of gas-phase axial mean $\langle U_z \rangle_g$ and r.m.s. fluctuations $u_z'g/\langle U_z \rangle_g$ along the spray axis, is illustrated in Fig. 4.9 for the three flames studied. For all the flames, $\langle U_z \rangle_g$ gradually increases until $z = 60$ mm. As the flow is convected downstream, a mean gas acceleration in the center regions takes place due to turbulent diffusion of momentum from regions away from the center. The peak values of $\langle U_z \rangle_g$ for cases HII and HIII exhibit a somewhat smaller magnitude than case HI, reflecting the slower mixing rates in the center. The results of $u_z'g/\langle U_z \rangle_g$ indicate that turbulence levels in the centerline are substantially higher for flame HI.
4.3.6 Gas-phase temperature field

The radial profiles of the gas-phase temperature statistics at four axial locations are shown in Fig. 4.10. On the left-hand side, the gas mean temperature $\langle T \rangle$ is presented along with the corresponding mean temperature gradient $\langle dT/dr \rangle$. On the right-hand side, the r.m.s. fluctuations $T'$ are presented.

At $z = 15$ mm, the $\langle T \rangle$ profiles show a rise from the coflow to a single peak centered around $r = 14$ mm for all cases. From the peak temperature location towards the spray axis, $\langle T \rangle$ gradually decreases to values lower than the ones observed in the coflow. However, in the region close to the spray centerline, $\langle T \rangle$ is significantly higher than the ethanol ignition temperature ($T_{ig} = 653$ K). The observed $\langle T \rangle$ profile shape can be attributed to the effects of evaporation and mixing prior to ignition as follows: due to the liquid atomization, an inward convective motion of the coflow is generated. The hot-diluted coflow is entrained into the spray and provides heat. The fuel vapor arises at the cost of heat losses from the oxidizing environment yielding lower gas-phase temperatures towards the spray axis. The mixture composition evolves from lean at the outer parts of the spray towards more rich at the center. Since the gas has relatively high temperature, the lean mixture at the spray outer parts ignites and the heat-release yields a peak in the $\langle T \rangle$. As one approaches the spray axis, the mean heat-release is lower and thus a decrease of $\langle T \rangle$ takes place. The peak $\langle T \rangle$ located at $r = 14$ mm is nearly identical for the three cases studied. This results from the combined effects of the coflow composition and temperature on the flame adiabatic temperature. The increase of the oxygen dilution in the oxidizer stream yields a reduction of the stoichiometric adiabatic temperature. However, a higher oxygen dilution in the coflow is accompanied by an increase of the overall temperature of the oxidizer stream. Thus, the two effects cancel out and no substantial differences among the three cases are observed.

At $z = 20$ mm, three observations can be made. First, for all cases, the peak $\langle T \rangle$ shifts towards larger radial distance and it is located in a region of large droplets and low droplet number density (see Fig. 4.7, $17 < r < 20$ mm). This effect results in a decrease of the mean temperature gradient from the peak $\langle T \rangle$ towards the center as compared to $z = 10$ mm. Secondly, the mean temperatures in the region $0 < r < 15$ mm are highest for case H1. This observation is in agreement with the sharper reduction of the droplet concentration in this region for case H1 (see Fig. 4.7). Lastly, throughout the flame region, the r.m.s. temperature fluctuation is rather similar among the three cases and the magnitude is overall higher by 75 K than in the hot-diluted coflow.

At higher axial stations, $z = 30$ and 40 mm, the displacement of the peak
4.3 Results and discussion

$\langle T \rangle$ towards larger radial distances proceeds due to the continuous spreading of the large droplets into the hot-diluted coflow. However, the distance between the peak and the spray axis is smaller for case H1. This indicates that spray flame H1 is narrower than the sprays flames in HII or HI. This is in agreement with the photographs presented in Fig. 4.3.

At $z = 40$ mm, a peculiarity in the radial profiles of $\langle T \rangle$ and $T'$ can be observed. This occurs more prominently in cases HII and HI and, therefore, a more detailed description is given. For these cases, between the radial positions $r = 0$ and $r = 10$ mm, $T'$ increases to a local peak at $r = 10$ mm and then decreases to smaller values until $r = 15$ mm. Also, a significant change in the $\langle T \rangle$ slope takes place at $r = 10$ mm. An authors' early study describes the spray flame structure in air (case AII) and in hot-diluted coflow (case HII). In air coflow, an inner and an outer flame-front are present starting from the lift-off position and diverging with increasing downstream location [44]. In the case of hot-diluted coflow (case HII), the inner flame-front is weak. Due to the local mixture richness the heat-release is significantly smaller as compared to the outer flame-front and a local maxima in $\langle T \rangle$ is not observed. The results of Fig. 4.10 show that for different coflow temperature and oxygen dilution an inner flame-front exists in all cases. It would be worthwhile to investigate this in more detail using other laser diagnostics such as OH-Planar Laser Induced Fluorescence.

Fig. 4.10 shows additional information concerning the influence of air entrainment on the coflow inlet temperature profiles. It can be noticed that the coflow temperature at $r = 40$ mm remains nearly the same throughout the axial stations presented. This shows that, for the range of radial distances displayed, the burner pipe diameter is wide enough to isolate the spray flame from the laboratory air entrainment. CARS measurements at higher axial stations (not shown) indicate that above $z = 60$ mm the outer region of the spray flame is perturbed by the laboratory air. Thus, below $z = 60$ mm, the spray flame is stabilized only by the hot-diluted coflow. Above the axial station of 60 mm, the oxidizer stream consists of a mixture of laboratory air together with hot-diluted coflow and reaction zone weakening effects may be present [81].
Figure 4.10: Gas-phase mean temperatures $\langle T \rangle$ (left) and r.m.s. fluctuations $T'$ (right) at four axial stations. The dashed grey lines correspond to the corresponding mean temperature gradients.
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4.3.7 Gas-phase temperature histograms

The normalized temperature histograms of cases H₁, H₁I and H₁II at z = 40 mm are presented in Fig. 4.11. Five radial positions were selected for each case. The histogram corresponding to the radial position in the coflow core region is represented by a thicker line. The remaining positions correspond to the spray region. A bin size of 50 K was selected for all the histograms presented. The total number of samples in each histogram is 1000, except for r = 0 mm of case H₁ where the number of valid samples was 120. For each of the five radial positions selected, the corresponding mean \( \langle T \rangle \), root-mean-square temperature fluctuation \( T' \) as well as skewness (S) and kurtosis (K) of the temperature distribution are listed in the table inset.

At the radial positions where the peak \( \langle T \rangle \) is observed (r = 20, 24 and 24 mm for cases H₁, H₁I and H₁II, respectively), the temperature distribution are nearly symmetric (S = 0) and broad with temperatures ranging from 2400 K down to 1400 K. A considerable amount of samples are over 2000 K for all cases. It is observed that the number of valid samples above 2000 K is greater for case H₁I. This is related to the higher temperature of the oxidizer coflow.

For all cases, the radial position of 15 mm is located between the inner and outer flame-front. As expected, lower temperature samples are present in the temperature distributions. As compared to the radial positions where the peak \( \langle T \rangle \) occurs, a substantial reduction of the number of valid samples above 2000 K is observed. For case H₁, a substantial amount of valid samples above 2000 K still persist at this position, yet, it is significantly reduced as compared to r = 20 mm. These results suggest that the coflow temperature is the driving parameter with respect to the outer flame-front temperature peaks.

At the spray axis, r = 0 mm, the temperature distributions are symmetric and samples are below 1200 K for all cases. Temperature histograms at z = 20 mm and 30 mm (not shown) have virtually identical r.m.s. and skewness value. This indicates that in the region close to the spray axis only turbulent mixing occurs and the gas-phase is rather homogeneous.
Figure 4.11: Normalized temperature histograms for five radial positions at $z = 40$ mm for cases $H_I$, $H_{II}$ and $H_{III}$. 
4.3.8 Droplet-size class properties

Fig. 4.12 and Fig. 4.13 show the mean axial $\langle U_z \rangle$ and radial $\langle U_r \rangle$ velocity components of four droplet size-classes at three axial stations, $z = 10$, 20 and 30 mm. The class width was chosen as 10 µm. All the points are an average of at least 200 samples. The droplets in the size class between 0 and 6 µm are considered to be flow tracers. The mean velocity of this size class thus represents the gas-phase mean velocity. The difference between the mean velocity for a particular size class and the gas-phase mean velocity will be referred to as slip velocity.

![Radial profiles of conditioned droplet mean axial velocity](image)

Figure 4.12: Radial profiles of the conditioned droplet mean axial velocity $\langle U_z \rangle$ for three axial stations. (●●● : Gas-Phase ($d \leq 6\mu m$), □□□ : $10\mu m \leq d < 20\mu m$, ○○○ : $20\mu m \leq d < 30\mu m$, △△△ : $30\mu m \leq d < 40\mu m$, + + + : $40\mu m \leq d < 50\mu m$)

The development of droplet $\langle U_z \rangle$ along the several axial stations shows similar trends for cases $H_I$, $H_{II}$ and $H_{III}$. At $z = 10$ mm, an increase of droplet
slip velocity with the size-class is observed. Farther downstream, slip velocity gradually decreases for the different droplet size-classes. Identical observations can be made regarding the droplet \( \langle U_r \rangle \) component. This occurs because the momentum of the liquid droplets is transferred to the gaseous phase through drag and evaporation.
4.3 Results and discussion

4.3.9 Discussion of the flame structure and lift-off behavior

This section presents a discussion on the droplet dispersion, flame structure and lift-off behavior of spray flame in hot-diluted coflow conditions. Before proceeding with the analysis, it is of interest to discuss in detail relevant dimensionless numbers characterizing the droplet motion.

Relevant dimensionless numbers

Some relevant dimensionless numbers are listed in Table 4.3 for each of the size classes and for the droplets smaller than 6 µm that are representative of the statistics of the gas-phase. The droplet Weber number (We$_d$) was determined as \( \rho_g U_j^2 d_c / \sigma \). Here, \( \rho_g \) represents the gaseous phase density, \( d_c \) is the mean droplet diameter of the selected size class and \( U_j \) the difference between the maximum droplet \( \langle U_z \rangle \) measured at \( z = 10 \) mm and the bulk coflow velocity. The gas properties were determined from the coflow bulk temperature and composition. As can be observed in Table 4.3, We$_d$ is well below unity for all droplet size classes suggesting that small droplets resulting from the secondary breakup are absent for all five cases.

The droplet Reynolds number (Re$_d$) was determined as \( U_j d_c / \nu_g \). Assuming that droplet are spherical and move in a viscous flow at a very low Reynolds number, the relaxation time \( (\tau_d) \) can be determined as \( \rho_d d_d^2 / 18\mu_g \). Here, \( \rho_d \) and \( \mu_g \) are the droplet density and the gaseous phase dynamic viscosity, respectively. As shown in Table 4.3, for droplets below 10 µm the Re$_d$ is well below one whereas for droplets bigger than 30 µm the highest value encountered for Re$_d$ is of 5. Hence, larger droplets incur an underestimation of the droplet relaxation time by at most 7%. On the other hand, droplets with a diameter smaller than 10 µm satisfy the assumption of Stokes flow.

The Stokes number (St$_d$) is defined as the ratio of the characteristic time of a droplet to a characteristic time of the flow and indicates the different regimes of droplet dispersion for each droplet size class [3, 51]. The flow characteristic time was defined as \( d_{at} / U_{at} \) where \( d_{at} \) is the atomizer nozzle diameter and \( U_{at} \) the estimated liquid jet velocity in the atomizer nozzle. Table 4.3 shows that for all cases studied, droplets bigger than 10 µm have St$_d$$\gg$1 and therefore do not follow changes in the flow field. For droplets with diameter smaller than 6 µm, the droplet response time is faster than the flow characteristic time scale and, therefore, behave as flow tracers.
Table 4.3: Relevant dimensionless numbers and relaxation time scales for different droplet size classes.

<table>
<thead>
<tr>
<th>Size-class</th>
<th>Cases</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>$H_1$</td>
</tr>
<tr>
<td>$We_d$ [-]</td>
<td></td>
</tr>
<tr>
<td>10-20 µm</td>
<td>0.0</td>
</tr>
<tr>
<td>20-30 µm</td>
<td>0.1</td>
</tr>
<tr>
<td>30-40 µm</td>
<td>0.2</td>
</tr>
<tr>
<td>40-50 µm</td>
<td>0.2</td>
</tr>
<tr>
<td>$Re_d$ [-]</td>
<td></td>
</tr>
<tr>
<td>10-20 µm</td>
<td>0</td>
</tr>
<tr>
<td>20-30 µm</td>
<td>1</td>
</tr>
<tr>
<td>30-40 µm</td>
<td>2</td>
</tr>
<tr>
<td>40-50 µm</td>
<td>3</td>
</tr>
<tr>
<td>$St_d$ [-]</td>
<td></td>
</tr>
<tr>
<td>0-6 µm</td>
<td>0.5</td>
</tr>
<tr>
<td>10-20 µm</td>
<td>11</td>
</tr>
<tr>
<td>20-30 µm</td>
<td>32</td>
</tr>
<tr>
<td>30-40 µm</td>
<td>62</td>
</tr>
<tr>
<td>40-50 µm</td>
<td>102</td>
</tr>
<tr>
<td>$\tau_d$ [ms]</td>
<td></td>
</tr>
<tr>
<td>0-6 µm</td>
<td>0.01</td>
</tr>
<tr>
<td>10-20 µm</td>
<td>0.17</td>
</tr>
<tr>
<td>20-30 µm</td>
<td>0.47</td>
</tr>
<tr>
<td>30-40 µm</td>
<td>0.93</td>
</tr>
<tr>
<td>40-50 µm</td>
<td>1.54</td>
</tr>
</tbody>
</table>

Flame structure

Fig. 4.14 presents a schematic of the spray structure in hot-diluted coflow conditions and, together with Table 4.3, can be used to describe the droplet dispersion and the flame structure. As a result of the spray breakup, droplets spread outwards and an outer flame-front develops wherein low droplet concentration and large droplets prevail. The outward ballistic motion of the droplets results from the droplet initial radial velocity as well as the correspondent large $St$ values and is the mechanism responsible for the flame inverted conical shape. As shown in Table 4.3, droplets bigger than 10 µm have Stokes
number sufficiently large to not follow changes in the flow field. Thus, as the spray evolves downstream, large droplets move outwards and a flammable mixture resulting from the entrained hot oxidizing coflow and the fuel vapor is formed at increasing larger radial distances. A comparison of cases H_I and H_III additionally shows highest Stokes number for droplets within the same size class when the coflow temperature is lowest (H_III). Hence, lower coflow temperatures yield a deeper penetration of droplets into the hot-diluted coflow and, consequently, the flames become wider. This is in agreement with the visual observations, namely the flame of case H_III is slightly wider than that of H_I (see Fig. 4.3 insets).

Figure 4.14: Schematic of the gas and droplet motion of pressure-swirl sprays in hot-diluted coflow conditions.
Characteristic scales and lift-off behavior

The trends presented in Fig. 4.4 show that the lift-off height decreases for increasing bulk coflow temperatures while it increases with increasing liquid mass flow rates. This section presents a possible explanation of the lift-off behavior of spray flames in hot-diluted coflow. As proposed by Delabroy et al. [71] and D. Martínez-Ruiz et al. [26], in a reacting spray, the time needed for combustion to take place is determined by the time scales of three phenomena: (1) droplet vaporization, (2) droplet convection and, (3) the chemical reaction. The comparison of the different time scales allows to characterize the lift-off height behavior of the spray flame. Estimates of characteristic numbers for different droplets size classes are presented in Table 4.4. The following discussion is based on single droplet theory and the interdroplet interaction effects are neglected [57, 69]. For all the calculations, the gas properties were determined from the coflow bulk temperature and composition.

The droplet vaporization time scale ($t_{vap}$) was estimated by the $d^2$ law [23, 59]. It is considered that droplets emerge from the atomizer nozzle at the boiling point and, therefore, a droplet heat-up period is absent. For an evaporating spherical droplet in a flow, $t_{vap}$ can be written as $d_0^2/K$. $K$ is the vaporization constant and is given by $-4/\rho_l[D_m Sh_m \ln(1 + B_M)]$. This model accounts for the slip velocity between the gas and droplet motion. The gas-phase properties values $'m'$ were evaluated by the 1/3-rule [100]. $D$ is the binary diffusion coefficient of ethanol and air at condition ($T_m$, $Y_m$). $Sh$ is the Sherwood number and $B_M$ represents the Spalding mass transfer number. Immediately after the atomizer nozzle exit plane, the droplets travel a certain path and leave behind a wake of fuel vapor that depends on the vaporization rates. Since the chemical reaction cannot begin until the gaseous fuel is generated and mixed, the vaporization time ($t_{vap}$) together with droplet convective time scale ($t_{conv}$) are two important scales influencing the lift-off location. An estimation of $t_{conv}$ was computed as $z_{10mm} / U_{zp}$. Here, $U_{zp}$ represents the velocity different between measured droplet $\langle U_z \rangle$ at the spray periphery at $z$ =10 mm and $U_{cf}$. A characteristic chemical time scale ($t_{ch}$) for the ethanol flame in hot coflow conditions was evaluated using calculations of transient counterflow diffusion flames. The strain rate was set to a representative value of $100s^{-1}$. The calculation started from the profile of a steady non-reaction flamelet (mixing only) and the chemical time $t_{ch}$ was defined as the time needed to reach the moment of maximal rate of temperature increase. The Chem1D code was used [22] using the Marinov chemical mechanism [65]. For the three cases, fuel vapor at the boiling temperature was used as fuel and the hot-diluted coflow as oxidizer. The evolution of the temperature as
4.3 Results and discussion

Function of time at the stoichiometric mixture fraction was used to estimate a characteristic chemical time scale for each of the three cases.

Table 4.4: Estimates of the characteristics time scales for different droplet size classes.

<table>
<thead>
<tr>
<th>Size-class</th>
<th>Cases</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>$H_I$</td>
</tr>
<tr>
<td>$t_{vap} \text{ [ms]}$</td>
<td></td>
</tr>
<tr>
<td>10-20 $\mu$m</td>
<td>0.21</td>
</tr>
<tr>
<td>20-30 $\mu$m</td>
<td>0.64</td>
</tr>
<tr>
<td>30-40 $\mu$m</td>
<td>1.21</td>
</tr>
<tr>
<td>40-50 $\mu$m</td>
<td>1.91</td>
</tr>
<tr>
<td>$t_{conv} \text{ [ms]}$</td>
<td></td>
</tr>
<tr>
<td>10-20 $\mu$m</td>
<td>3.47</td>
</tr>
<tr>
<td>20-30 $\mu$m</td>
<td>1.84</td>
</tr>
<tr>
<td>30-40 $\mu$m</td>
<td>1.20</td>
</tr>
<tr>
<td>40-50 $\mu$m</td>
<td>0.99</td>
</tr>
<tr>
<td>$t_{ch} \text{ [ms]}$</td>
<td>0.31</td>
</tr>
<tr>
<td>$t_{vap}/t_{conv} [-]$</td>
<td>0.06</td>
</tr>
<tr>
<td>20-30 $\mu$m</td>
<td>0.35</td>
</tr>
<tr>
<td>30-40 $\mu$m</td>
<td>1.01</td>
</tr>
<tr>
<td>40-50 $\mu$m</td>
<td>1.92</td>
</tr>
<tr>
<td>$t_{ch}/t_{vap} [-]$</td>
<td>1.48</td>
</tr>
<tr>
<td>20-30 $\mu$m</td>
<td>0.48</td>
</tr>
<tr>
<td>30-40 $\mu$m</td>
<td>0.26</td>
</tr>
<tr>
<td>40-50 $\mu$m</td>
<td>0.16</td>
</tr>
</tbody>
</table>

A comparison between cases $H_I$, $H_{II}$ and $H_{III}$ shows no substantial differences on the droplets convective time scale. However, significant differences are observed in the droplets vaporization and in the chemical time scales. For droplets smaller than 30 $\mu$m, $t_{vap}$ is lower than $t_{conv}$ by almost one order of magnitude. This indicates that substantial vaporization of droplets smaller than 30 $\mu$m occurs prior to the lift-off location. As expected, larger droplets survive the near-injector region due to their larger Stokes number and spread.
into the hot-diluted coflow.

For the highest coflow temperature (case $H_1$), droplets smaller than 10 µm have $t_{\text{vap}}$ of the same order of magnitude as that of $t_{\text{ch}}$. Furthermore, $t_{\text{vap}}$ and $t_{\text{ch}}$ are considerably smaller than $t_{\text{conv}}$. This indicates that chemical reaction engages almost immediately after the gaseous fuel is generated from the small droplets in the near-injector region and the vapor fuel availability is the limiting factor to form an ignitable mixture. The lower coflow bulk temperatures of cases $H_{II}$ and $H_{III}$ yield an increase of the $t_{\text{vap}}$ and, consequently, less fuel vapor is available prior to the lift-off location. A comparison between the different droplet size classes of case $H_1$ and $H_{III}$ show a reduction of about 60% in the vaporization time scales for droplets in the size class of 20-30 µm and of 30% for droplets in the size class of 40-50 µm (see bold values in Table 4.4). The chemical time scale increases by more than one order of magnitude and is noticeably bigger than $t_{\text{vap}}$ or $t_{\text{conv}}$. It thus can be understood that for case $H_{II}$ and $H_{III}$, both $t_{\text{vap}}$ and $t_{\text{ch}}$ contribute to an increase of the lift-off height, however, $t_{\text{ch}}$ has a more prominent effect.

The impact of the liquid mass flows on the lift-off height can be assessed by comparing case $H_1$ with $H_{I}^{\text{aux}}$ and $H_{II}$ with $H_{II}^{\text{aux}}$. Increase of the liquid mass flows (and, therefore, higher liquid jet momentum) yields a decrease of $t_{\text{conv}}$ due to the higher droplet initial velocity. However, only a slight change is observed in the vaporization rates. Hence, the concentration of fuel vapor in the droplets wake until $z = 10$ mm is smaller and, consequently, a larger droplet path is required to form a flammable mixture. This manifests itself as an increase of the lift-off height.
4.4 Conclusions

A comprehensive study was carried out of ethanol sprays issuing into hot-diluted coflows with different conditions. LDA, PDA and CARS techniques were employed to study spray flame structure and provide an explanation of the stabilization characteristics of sprays combustion under Mild conditions. An extensive database was obtained for validation of numerical simulations.

For pressure-swirl sprays with identical Weber number the results indicate that within the coflow conditions studied the initial droplet velocity and $d_{32}$ distribution do not differ significantly. Small droplets are present at the center and quickly evaporate as the spray evolves downstream. Large droplets with relatively high Stokes number move outwards and penetrate into the hot-diluted coflow. The fuel vapor originated from the large droplets mixes with the hot-diluted coflow and a lean flame-front develops in a region of low droplet concentration at the spray outer parts. The temperature histograms in this region show a considerable amount of samples over 2000 K. Lower coflow bulk temperature yields a reduction of the peak temperatures in the lean flame-front at the spray outer parts suggesting that the oxidizer stream temperature is the driving parameter with respect to the peak temperatures. Due to the initial droplet concentration distribution, the gas composition becomes increasingly rich towards the spray axis. A secondary flame-front significantly weakened due to the local richness of the gas composition is also observed close to the spray center region for all the cases studied. It is shown that the observed increase of the lift-off height for lower coflow bulk temperatures is related to the increase of the droplet vaporization time scale and an increase of the chemical time scale.
CHAPTER 5

Ethanol and acetone spray flames in hot-diluted coflow conditions

This chapter presents measurements of an ethanol and an acetone turbulent spray flame issuing into a coflow of 10.1% oxygen volume fraction at 1200 K. The liquid mass flow rates were chosen to yield spray flames with identical Weber number. High-speed visualizations were performed to ascertain the atomization mechanisms of ethanol and acetone liquid fuels in hot-diluted conditions. Laser Doppler anemometry, phase Doppler anemometry and coherent anti-Stokes Raman spectroscopy were applied in the spray flame and the coflow to provide a thorough description of the sprays structure as well as corresponding inflow boundary conditions.

High-speed visualizations show that in the ethanol case, the liquid jet disintegrates into droplets and ligaments through perforations in a liquid conical sheet. This contrasts greatly with the acetone case wherein a liquid-vapor mixture is issued from the atomizer nozzle due to presence of cavitation inside the atomizer. The visual appearance of the two spray flames is found to differ drastically with the acetone spray flame lift-off height higher than the ethanol spray flame. It is observed that droplet convective time scales plays a key role on the lift-off location. As a result of cavitation, droplet convective time scales are substantially reduced leading to an increase of the axial location where a ignitable mixture is formed. In the downstream region of the ethanol spray case, wherein droplets are already evaporated, a substantial amount of temperature samples above the expected adiabatic temperature are observed. This observation is attributed to the formation of H₂.
5.1 Cases description

The burner facility has been described in detail in Chapter 2. In brief, the burner consists of a pressure-swirl spray within an annular coflow of exhaust products from a secondary burner. Both the liquid jet and coflow conditions are controlled separately. One ethanol (H_{III}) and one acetone (AcH_{III}) spray flame in hot-diluted coflow are studied. The bulk temperature and oxygen concentration in the coflow are identical for both cases. The properties of the liquid fuels serve as background information for the following discussion and are presented in Table 5.1. The burner operating conditions and the relevant dimensionless parameters of each spray flame are given in Table 5.2.

$U_{cf}$, $T_{cf}$ and $(X_{O_2})$ represents the mass average velocity, temperature and oxygen volume fraction of the coflow, respectively. The values were determined by the measured velocity, temperature and oxygen volume fraction profiles at $z = 0$ mm, i.e. at the atomizer orifice exit plane. The Reynolds number of the coflow ($Re_{cf}$) was based on the hydraulic pipe diameter ($D_h = 140$ mm) and the thermophysical properties of the gas estimated from the coflow bulk temperature. The Weber number is defined as $\rho_g U_{jet}^2 l/\sigma$. $l$ is the characteristic liquid film thickness and it was assumed as equal to the nozzle diameter, i.e. 210 µm. $U_{jet}$ represents the difference between the estimated liquid jet velocity from the liquid mass flow rate and $U_{cf}$. The liquid mass flow rates were selected to yield similar Weber number for the two cases. The positions where the measurement techniques were employed are summarized in Table 5.3.

The velocity profiles at $z = 0$ mm are displayed on Fig. 5.1 for cases H_{III} and AcH_{III}. On the left-hand side, the coflow axial mean velocity $(U_z)$ and the corresponding r.m.s. fluctuations $u'_z$ are presented. The same quantities are shown for the radial velocity component on the right-hand side.

Some differences can be observed in the near-wall region for the two cases. For case H_{III}, $(U_z)$ exhibits a sharp increase between $r = 10$ and 11 mm which

<table>
<thead>
<tr>
<th>Fuel</th>
<th>Density [kg/m$^3$]</th>
<th>Surface tension [mN/m]</th>
<th>Kinematic viscosity $\times 10^{-6}$ [m$^2$/s]</th>
<th>Vapor pressure [kPa]</th>
<th>Boiling point [K]</th>
<th>Enthalpy of vaporization [kJ/kg]</th>
<th>Ignition temperature [K]</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ethanol</td>
<td>785</td>
<td>21.78</td>
<td>1.39</td>
<td>7.883</td>
<td>351</td>
<td>919</td>
<td>638</td>
</tr>
<tr>
<td>Acetone</td>
<td>781</td>
<td>22.16</td>
<td>0.38</td>
<td>30.397</td>
<td>329</td>
<td>539</td>
<td>738</td>
</tr>
</tbody>
</table>
Table 5.2: Mass flows for the coflow and spray are presented for the studied test flames. The dimensionless numbers as well as coflow mass-averaged quantities are also presented. The pressure fluctuations are less than 0.2% for all flames.

<table>
<thead>
<tr>
<th>Case</th>
<th>$\dot{m}_{\text{air}}$ [kg/h]</th>
<th>$\dot{m}_{\text{DNG}}$ [kg/h]</th>
<th>$U_{\text{ef}}$ [m/s]</th>
<th>$T_{\text{ef}}$ [K]</th>
<th>$\langle X_{O_2} \rangle$ [-]</th>
<th>$\dot{m}_{\text{Liq}}$ [kg/h]</th>
<th>$p$ [bar]</th>
<th>$T_{\text{Liq}}$ [K]</th>
<th>$U_{\text{jet}}$ [m/s]</th>
<th>$W_e$ [-]</th>
<th>$\xi_{st}$ [-]</th>
</tr>
</thead>
<tbody>
<tr>
<td>H_I</td>
<td>45</td>
<td>1.75</td>
<td>1.81</td>
<td>1225</td>
<td>10.1</td>
<td>1.48</td>
<td>11.7</td>
<td>25</td>
<td>15.1</td>
<td>0.4</td>
<td>0.052</td>
</tr>
<tr>
<td>AcH_I</td>
<td>1.15</td>
<td>11.7</td>
<td>25</td>
<td>12.3</td>
<td>0.5</td>
<td>1.15</td>
<td>11.7</td>
<td>25</td>
<td>12.3</td>
<td>0.5</td>
<td>0.046</td>
</tr>
</tbody>
</table>

Table 5.3: Summary of measurement locations for the two flames studied.

<table>
<thead>
<tr>
<th>Measurement techniques:</th>
<th>z [mm]</th>
<th>H_I</th>
<th>AcH_I</th>
<th>Comment:</th>
</tr>
</thead>
<tbody>
<tr>
<td>LDA</td>
<td>0, 20, 25, 30, 35, 40, 45</td>
<td>0</td>
<td>Centerline, Coflow radial profiles</td>
<td></td>
</tr>
<tr>
<td>CARS</td>
<td>0, 15, 20, 30, 40, 50, 60</td>
<td>0, 20, 30, 40, 50, 60</td>
<td>Coflow radial profiles, Spray region</td>
<td></td>
</tr>
<tr>
<td>Flue gas analyser</td>
<td>0</td>
<td>0</td>
<td>Coflow radial profiles</td>
<td></td>
</tr>
<tr>
<td>PDA</td>
<td>8, 10, 12, 15, 20, 30, 35, 40, 45</td>
<td>8, 10, 12, 15, 20, 30, 35, 40, 45</td>
<td>Spray region, Steps of 1 mm</td>
<td></td>
</tr>
</tbody>
</table>
is followed by a decrease until a region with fairly constant values. For case AcHIII, the increase of \( \langle U_z \rangle \) in the near-wall region is smaller as compared to HIII. On opposite sides of the spray, for both cases the magnitude of \( \langle U_r \rangle \) increases until a maximum in the near-wall of the central rod and then gradually decreases outwards. This observation indicates that the coflow has a strong inward velocity component and is being convected towards the spray axis. This effect has been shown previously to result from the presence of the spray [44] and is more pronounced for case AcHIII.

Fig. 5.2 displays the coflow mean temperature \( \langle T \rangle \), r.m.s. fluctuations \( T' \) and oxygen volume fraction \( \langle X_{O_2} \rangle \) at \( z = 0 \) mm for cases HIII and AcHIII. Fig. 5.2 show that \( \langle X_{O_2} \rangle \) radial profile is fairly uniform across the coflow. In the \( \langle T \rangle \) profiles, \( \langle T \rangle \) decreases towards the fuel rod wall due to the heat losses to the water-cooling system. Asymmetries are seen that exceed the inaccuracy of the CARS system. The maximum difference between \( \langle T \rangle \) of corresponding positions at opposite sides of the axis amounts to 43 K.
5.2 Results and discussion

This section presents the measurement results of the ethanol and acetone spray flame cases. At first, a description of the flames visual appearance and lift-off height observations is presented for both cases. The proceeding discussion concerns the liquid jet breakup and the most notable features of the spray flames structure for each individual case. The last section ties together the experimental results and discusses the implications of the findings to the understanding of MILD combustion of different liquid fuels.

5.2.1 Visual appearance of the spray flames - $H_{III}$ and $AcH_{III}$

To show the overall flame appearance, color pictures are displayed in Fig. 5.3. The flames were photographed using a digital camera (Canon EOS X5) with 0.6 s exposure time and identical aperture. The inset shows a close-up of the spray region and initial development of the flame. This is the region of

Figure 5.2: Mean oxygen volume fraction $\langle X_{O_2} \rangle$, mean temperature $\langle T \rangle$ and root-mean-square temperature fluctuations $T'$ profiles at $z = 0$ mm for cases $H_{III}$ and $AcH_{III}$. 
Ethanol and acetone spray flames in hot-diluted coflow

Figure 5.3: Images of test cases with 0.6s exposure time and identical aperture. The field-of-view of the inset are equal for both cases and has dimensions $180 \times 160 \text{mm}^2$ (width $\times$ height).

Three regions can be distinguished in the case H$_{\text{III}}$ from its visual appearance: a jet liquid breakup region, a faint bluish reaction zone and faint yellowish zone farther downstream. The flame is weakly luminescent, transparent and relatively bright in the outer-regions of the spray. Case AcH$_{\text{III}}$ displays distinct features from that of case H$_{\text{III}}$. As can be observed in the inset, the lift-off height is at a higher axial location and no bluish reaction zone is observed. This is contrary to what would be predicted based on the comparison of enthalpy of vaporization and boiling point of the two liquid fuels (see Table 5.1). This observation of is related to the chemical time scales of the fuel and will be discussed in further detail in Section 5.2.3. In the far downstream region a yellow-zone with significantly stronger luminescence than in case H$_{\text{III}}$ is observed.
5.2 Results and discussion

5.2.2 Ethanol spray flame in hot-dilute coflow - Case H_{III}

Liquid jet breakup

High-speed visualizations of the liquid breakup were performed for case H_{III}. Six sequential images are displayed in fig. 5.4. Exposure time and camera frame rate was adjusted to fully resolve the liquid disintegration and to produce an optimal optical contrast. The frame rate and field-of-view selected was 125 kHz and $6.81 \times 3.04 \text{ mm}^2$, respectively. The resolution of the imaging system was 26.6 µm per pixel.

From the sequential images, it is observed that ethanol emerges from the atomizer orifice as a swirling liquid sheet (see fig. 5.4a). The continuous length of the liquid jet prior to breakup is about 1 mm. On the liquid jet surface, small oscillations are observed. As the liquid jet proceeds away from the atomizer exit, small disruptions take place on the liquid jet surface (see fig. 5.4d) and the liquid jet quickly disintegrates into droplets and ligaments (see fig. 5.4e).

In chapter Chapter 3 it is shown that large- and small-perturbations on the jet surface occur and may grow as a result of the competition of cohesive and disruptive forces. When the contribution of the forces generating instability in the liquid jet are higher than the cohesive forces, the perturbations may undergo further wave growth depending on the interaction of the liquid surface with surrounding air flow. These growing perturbations lead to a local thinning of the conical liquid sheet and, subsequently, to liquid jet disruption. In the presence of hot-diluted coflow, it is hypothesized that vaporization effectively contributes to the local thinning of the liquid sheet and, therefore, a wider classe of perturbations leads to a disintegration of the liquid sheet into ligaments and droplets. It can be argued that the implosion of cavitation bubbles formed inside the atomizer may initiate the liquid disintegration. However, no evidences of this are observed in the high-speed visualizations.

Droplet Diameter distribution and Mean Velocity Fields

Profiles of $d_{32}$ at five axial distances, $z = 10, 15, 20, 30$ and 40 mm along with the mean gas and droplet velocity fields are presented in Fig. 5.5 for case H_{III}. The dashed line superimposed in the mean velocity field marks the locations where flame luminescence reaches a maximum.

At $z = 10$ mm, droplets have small $d_{32}$ in the center-region and move mainly upwards. At larger distances from the spray axis, the $d_{32}$ increases until a maximum. Along with the $d_{32}$, also the average radial velocity increases. This indicates that a spreading of the spray into the hot-diluted coflow occurs and agrees with the visual observations. Above $z = 15$ mm, flame luminescence
Figure 5.4: Sequence of images showing the liquid sheet breakup for case $H_{III}$ ($We = 0.4$). The field-of-view has dimensions $6.81 \times 3.04 \text{ mm}^2$. 
Figure 5.5: Sauter mean diameter \( (d_{32}) \) profiles and mean gas and droplet velocity fields for case H\(_{III} \). The dashed lines marks the maximum flame luminescence.

...starts to be visible. A substantial portion in the center of the spray becomes void of droplets and large droplets appear in a region away from the flame luminescence. As the spray proceeds downstream, evaporation and dispersion into the hot-diluted coflow continues to occur and the flame luminescence is displaced radially outwards. No droplets are observed for axial locations above \( z = 45 \text{ mm} \).
Gas-phase mean velocity, temperature and droplet number density

To study the influence of the droplets on the local turbulence structure and composition of the gas-phase, droplet number density $n_d$ along with the gas-phase mean axial velocity $\langle U_z \rangle_g$ and mean temperature $\langle T \rangle$ radial profiles are plotted in Fig. 5.6. These quantities are presented at four axial stations, $z = 10, 15, 20$ and 35 mm.

To represent the large amount of data, the physical properties are shown together in one figure. The differently colored $y$-axis scales are used for the aforementioned physical quantities and the corresponding ranges have been preserved over the four axial stations. Droplets with diameter below $6 \mu m$ from the full record were taken as gas tracers. The results of the gas-phase are limited by the availability of the small droplets below $6 \mu m$ and each point presented in Fig. 5.6 is based on an ensemble of, at least, 100 samples. The datatips displayed in the figure indicate the positions where the maximum flame luminescence is observed. Fig. 5.7 displays the radial distribution of the root-mean-square temperature fluctuations $T'$ and complements the information given in Fig. 5.6. The position of the flame luminescence at a given axial station is identified in this figure by the red squares.

In Fig. 5.6, it can be observed that at $z = 10$ mm droplets are mainly concentrated around $r = 7$ mm and $\langle U_z \rangle_g$ profiles exhibit a 'dip' in the spray axis. From $z = 10$ mm to $z = 15$ mm, an increase of $\langle U_z \rangle_g$ along the spray centerline takes place due to the turbulent diffusion of momentum. Also, a temperature rise of about 500 K from the coflow core to a single peak occurs at $z = 15$ mm, $r = 13$ mm. The $\langle T \rangle$ gradually decreases from the peak temperature towards the spray axis where the observed values are lower than that of the coflow, yet, significantly higher than the ethanol ignition temperature ($T_{ig} = 653$ K). This results from the temperature and fuel vapor distribution in the spray region prior to ignition. At the spray most outer parts, the mixture results from the fuel-vapor of the droplets located at the spray most outer parts and the coflow entrainment. This yields a lean mixture where the oxidation process takes place. At positions closer to the center axis, the local mixture composition becomes increasingly rich and the heat-release is gradually smaller. As the spray evolves downstream, droplets penetrate further into the hot-diluted coflow due to their initial outward radial momentum and larger Stokes number. As consequence of the presence of liquid fuel at larger radial distances, the peak $\langle T \rangle$ moves towards larger radial distances.

In Fig. 5.7 an additional observation can be made. At $z = 30$ mm, $T'$ increases to a local peak at $r = 10$ mm. Also, at the same radial position, a local change in the $\langle T \rangle$ decay slope takes place (see Fig. 5.6). This suggests that a
Figure 5.6: Radial profiles of gas-phase mean axial velocity $\langle U_z \rangle_g$, mean temperature $\langle T \rangle$ and droplet number density $n_d$ at four axial stations for case $H_{III}$.
flame-front is also present inside the spray region. However, the heat-release is substantially smaller as compared to the flame-front at the spray most outer parts. Furthermore, it suggests that within $0 < r < 10 \text{ mm}$ the mixture is too rich to engage in the oxidation process and only mixing occurs.

Gas-phase Reynolds stresses

Gas-phase normal $\langle u_z u_z \rangle_g$, radial $\langle u_r u_r \rangle_g$ and shear $\langle u_z u_r \rangle_g$ Reynolds stress profiles at four axial stations are shown in Fig. 5.8 for case H$_{\text{III}}$. The axial stations presented correspond to the region where a substantial number of droplets is present. Here, the degree of anisotropy of the turbulence in the gas flow is characterized by the ratio between the normal and radial Reynolds stresses, i.e. $\langle u_z u_z \rangle_g / \langle u_r u_r \rangle_g$.

Fig. 5.8 shows that $\langle u_z u_z \rangle_g$ is substantially higher than $\langle u_r u_r \rangle_g$ at $z = 10 \text{ mm}$ indicating that a strong anisotropy and significant mixing takes place near the atomizer region. As the gas-phase evolves downstream, a fast decay of fluctuations levels is observed resulting in a nearly isotropic flow in the regions farther downstream.
5.2 Results and discussion

Figure 5.8: Gas-phase normal $\langle u_z u_z \rangle_g$, radial $\langle u_r u_r \rangle_g$ and shear $\langle u_z u_r \rangle_g$ Reynolds stresses profiles at $z = 10, 15, 20, 30$ mm for case H_{III}.

Droplet-size class properties

To illustrate the characteristics of the droplets behavior, the mean axial $\langle U_z \rangle$ and radial $\langle U_r \rangle$ velocity profiles of four droplet size-classes at three axial stations, $z = 10, 20$ and $30$ mm, are shown in Fig. 5.9. The class width was chosen as $10 \mu$m. All the points are an average of at least 200 samples. The mean velocity of the droplets in size class $0 - 6 \mu$m are considered to be flow tracers and thus represent the gas velocity. Hereafter, the difference between the mean velocity for a particular size class and the gas-phase mean velocity is referred to as ‘slip velocity’.
Figure 5.9: Mean axial $\langle U_z \rangle$ and radial $\langle U_r \rangle$ velocities profiles per size class for case H$_{III}$.

The $\langle U_z \rangle$ and $\langle U_r \rangle$ profiles of the different size-classes show an increase of the droplet slip velocity with increasing droplet diameter. Farther downstream, a gradual reduction of the slip velocity for all the size classes is observed. This occurs because the momentum of the liquid droplets is transferred to the gaseous gas and, consequently, causes the droplets to reduce their initial velocity. At $z = 30$ mm the different size-class radial profiles exhibit a similar shape mean axial and radial velocity profiles.
Gas-phase temperature evolution in the downstream region

For axial stations above $z = 40$ mm, droplets are absent and no gas-phase statistics can be retrieved from the small droplets. This is the region where the yellowish faint-brush develops. CARS measurements were performed to unveil the gas-phase temperature evolution.

The results are shown in Fig. 5.10 for $z = 30, 40, 50$ and $60$ mm. On the left-hand side, the gas-phase mean temperature $\langle T \rangle$ is presented along with the corresponding mean temperature gradients $(d\langle T \rangle/dr)$. On the right-hand side, the r.m.s. fluctuations $T'$ are presented.

From $z = 30$ mm to $z = 40$ mm, the peak $\langle T \rangle$ shifts from $r = 21$ mm to $24$ mm. This can be explained by the deeper penetration of the large droplets into the hot-coflow. As the fuel droplets more outwards, more fuel vapor is available at large radial distances and, therefore, the location where the stoichiometric mixture takes place is displaced towards larger radial distances. The mean temperature gradients $(d\langle T \rangle/dr)$ along the flame cross-section are reduced. Some peculiarities can be observed in the region farther downstream. From $z = 40$ mm to $50$ mm, the magnitude of the peak $\langle T \rangle$ increases substantially and is followed up by a reduction at $z = 60$ mm. Furthermore, the $T'$ slightly increases over the region where the oxidation process takes place.

The measurements in the hot-diluted coflow region, i.e. between $r = 40$ and $80$ mm, reveal where the influence of air entrainment in the spray flame starts to take place. The laboratory air gradually mixes with the hot-diluted coflow changing the coflow initial radial temperature and oxygen distribution. Up to $z = 60$ mm, the burner pipe diameter is wide enough to isolate the spray flame from the laboratory air entrainment. Thus, the flame is stabilized only by the hot-diluted coflow. Above the axial station of $60$ mm, the oxidizer stream consists of a mixture of laboratory air together with hot-diluted coflow and reaction zone weakening effects may be present [81].

Gas-phase temperature histograms

The normalized temperature histograms at $z = 40, 50$ and $60$ mm are presented in Fig. 5.11 for case HIII. The five radial positions were selected to evaluate the trends in the temperature histograms along the flame cross-section. A bin size of $50$ K was selected for all the histograms presented. The total number of samples in each histogram amounts to $1000$. For each of the five radial positions selected, the corresponding mean $\langle T \rangle$, root-mean-square temperature fluctuation $T'$ as well as skewness $(S)$ and kurtosis $(K)$ of the temperature distribution are presented in the inset. Skewness negative values indicate data
Figure 5.10: Gas-phase mean temperatures $\langle T \rangle$ (left) and r.m.s. fluctuations $T'$ (right) at four axial stations. The dashed grey lines correspond to the correspondent mean temperature gradients ($d\langle T \rangle/dr$).
that are skewed towards lower temperatures and that the left tail is long relative to the right tail. The high kurtosis values indicate that the distribution has a sharper peak and longer, fatter tails.

At \( z = 40 \text{ mm} \), \( r = 40 \text{ mm} \), \( \langle T \rangle \) and \( T' \) are 1314 K and 101 K, respectively. At the radial position where the peak \( \langle T \rangle \) is observed, \( r = 24 \text{ mm} \), the normalized temperature histogram shape are rather alike, yet, shifted towards higher temperatures. A few samples are above the ethanol adiabatic temperature \( (T = 2200 \text{ K}) \). At \( z = 50 \text{ mm} \), from the coflow core \( (r = 41 \text{ mm}) \) to the radial position where the peak \( \langle T \rangle \) is observed \( (r = 24 \text{ mm}) \), the normalized histograms exhibit the same qualitative behavior. However, a substantial amount of samples are above 2200 K. These results appear contrary to what is expected since a substantial amount of samples largely surpass the ethanol theoretical adiabatic temperature in stoichiometric conditions. At \( z = 60 \text{ mm} \), \( r = 27 \text{ mm} \), i.e. at the radial position where the peak \( \langle T \rangle \) is observed, the number of samples above 2200 K is greatly reduced compared to \( z = 50 \text{ mm} \), \( r = 24 \text{ mm} \).

In order to exclude the possibility of an experimental error, some additional verifications were undertaken to check the validity of the CARS measurements. Due to flame radiation, the optical components are subject to a substantial thermal load during the measurement campaigns. Consequently, the wavelength position and the shape of the excitation profile, may change during a series of measurements leading to systematic errors [34]. The \( \langle T \rangle \) profiles at \( z = 50 \text{ and } 60 \text{ mm} \) were obtained on the same measurement session. Therefore, in case this effect was present, systematic errors would be expected in the flame region as well as on the coflow wherein no droplets are observed at both axial stations. The difference of \( \langle T \rangle \) between the two axial stations at \( r = 41 \text{ mm} \) is of 8 K which is well below the CARS system inaccuracy. To further strengthen the validity of the CARS measurements, the CARS data was post-processed with different excitation profiles obtained during the measurement campaigns. The outcome show that a large number of temperature above 2200 K persist in the temperature histograms at \( z = 50 \text{ mm} \), \( r = 24 \text{ mm} \). This is the adequate proof that the observed variation is real and not a mere experimental error.

The appearance of the hotter samples can be attributed to the presence of finite rate kinetics and differential diffusion in the rich faint brush region downstream of the spray flame as described by Neophytou et al. [6]. Specifically, in regions where the oxidiser was completely consumed in the reaction zone and vapor fuel is pyrolysed, small species such as \( \text{C}_2\text{H}_2 \), \( \text{C}_2\text{H}_4 \) and \( \text{H}_2 \) may be formed and enhance the local reaction rates. The presence of \( \text{H}_2 \), thus, may lead to local peak temperatures above the ethanol adiabatic temperature.
Figure 5.11: Normalized temperature histograms for five radial positions at $z = 40$, 50 and 60 mm for case H_{III}. 
5.2 Results and discussion

5.2.3 Acetone spray flame in hot-diluted coflow - Case AcH_{III}

The present section concerns the results of the acetone flame (AcH_{III}) and are presented in the same manner as the ethanol flame (H_{III}). The most notable features of the ethanol flame are recalled whenever appropriate so as to compare with the distinct peculiarities of the acetone flame.

**Liquid breakup high-speed visualizations**

High-speed visualizations of the liquid breakup were also performed for case AcH_{III}. Six sequential images are displayed in Fig. 5.12. The frame rate selected for case AcH_{III} was 90 kHz. As a result, the field-of-view is larger than case H_{III} and has dimensions 10.21×3.04 mm\(^2\). The resolution of the imaging system was 26.6 µm per pixel and is equal to case H_{III}.

The sequence of images displayed in Fig. 5.12 show significant differences between cases AcH_{III} and H_{III}. In case AcH_{III}, a column of droplets immediately emerges from the atomizer nozzle and disperses into the surrounding hot-diluted environment. This observation can be attributed to cavitation inside the atomizer and demonstrates the different mechanism responsible for the liquid disintegration in both cases. Further downstream, the occurrence of coalescence and secondary breakup is also evident.

**Droplet Diameter distribution and Mean Velocity Fields**

Profiles of \(d_{32}\) at five axial distances, \(z = 10, 15, 20, 30, 40\) mm along with the mean gas and droplet velocity fields are presented in Fig. 5.13 for case AcH_{III}. The dashed line superimposed in the mean velocity field marks the locations where flame luminescence reaches a maximum.

There are substantial differences in terms of spray structure and droplet dispersion in the hot-diluted coflow as compared to H_{III} (see Fig. 5.5). As can be observed from the mean velocity fields, the spray is substantially narrower. Within the domain shown, both the droplets and mean gas flow field have a higher velocity as compared to case H_{III}. At \(z = 10\) mm, the droplet \(d_{32}\) along the spray cross-plane is highly non-uniformly distributed. Smaller \(d_{32}\) is observed near the centerline, yet, it is significantly higher than case H_{III}. This indicates that a substantial number of larger droplets is present in this region as compared to H_{III}. With increasing radial distance from the centerline, \(d_{32}\) increases until a maximum and then progressive decreases. As the spray evolves downstream, it is observed that droplets subsist in the center region as
Figure 5.12: Sequence of images showing the liquid sheet breakup for case AcH_{III} (We = 0.5). The field-of-view has dimensions $10.21 \times 3.04 \text{ mm}^2$. 
Figure 5.13: Sauter mean diameter ($d_{32}$) profiles and mean gas and droplet velocity fields for case Ac$H_{III}$. The dashed lines marks the maximum flame luminescence.

opposed to what is observed in case $H_{III}$. The $d_{32}$ increases at the spray most outer parts due to the evaporatization of the small droplets and the outward motion of the large droplets. A visible flame is observed at about $z = 45$ mm.
Gas-phase velocity, temperature and droplet number density

Fig. 5.14 shows the gas-phase mean axial velocity $\langle U_z \rangle_g$ and the mean temperature $\langle T \rangle$ along with droplet number density $n_d$ at four axial stations for case AcHI. Below $z = 20$ mm, severe droplet breakdown events were observed and a statistical number of CARS valid spectra was not achieved.

As compared to case HII, some notorious differences can be observed in the spray initial droplet density and gas-phase velocity distribution. At $z = 10$ mm, $n_d$ indicates that the liquid droplets are distributed uniformly in the region close to the centerline. Moreover, the droplets and the gas-phase axial velocity magnitude is substantially larger than that of case HII. This stems from the different liquid breakup mechanisms existent in case AcHI and HII. As shown in Fig. 5.4, ethanol fuel is issued from the atomizer nozzle as a liquid jet that swirls and disrupts through instabilities originated on the liquid sheet. In case AcHI however, the fuel is already issued from the atomizer nozzle as a mixture of fuel vapor and liquid. The portion of fuel vapor significantly reduces the mixture density and yields a substantial increase of the velocity. Moreover, droplets accelerate in the gas core and, subsequently, the ratio of the axial-to-radial velocity components is substantially larger reducing significantly the spray cone angle of case AcHI.

As the spray evolves downstream, the large droplets gradually disperse into the hot-diluted coflow. It is observed that $\langle T \rangle$ gradually decreases from the coflow core to lower values at the spray axis. This results from the heat exchange between the droplets and the entrained hot-diluted coflow and supports the hypothesis presented for case HII in Section 5.2.2. As the hot-diluted coflow is entrained towards the spray center, it encounters an increasing number of droplets and, therefore, available surface area for heat and mass transfer. A significant amount of energy is needed to overcome the liquid latent heat and vaporize the liquid droplets. Hence, the gas-phase temperature decreases from the coflow core to lower values at the spray axis. Surprisingly, although in case AcHI droplets are mainly concentrated in the center, the $\langle T \rangle$ observed along the centerline are slightly higher than that of case HII.

Gas-phase Reynolds stresses

Gas-phase normal $\langle u_z u_z \rangle_g$, radial $\langle u_z u_r \rangle_g$ and shear $\langle u_z u_r \rangle_g$ Reynolds stress profiles at four axial stations are shown in Fig. 5.16 for case AcHI. The scale has been kept identical to Fig. 5.8 so to allow a direct comparison between the two cases.

In the near atomizer region, $z = 10$ mm, Reynolds stresses are significantly
Figure 5.14: Radial profiles of gas-phase mean axial velocity $\langle U_z \rangle_g$, mean temperature $\langle T \rangle$ and droplet number density $n_d$ at four axial stations for case AcH$_{III}$. 
Figure 5.15: Root-mean-square temperature fluctuations $T'$ at several axial stations for case $\text{AcH}_{\text{III}}$. The red-square represented the location of the maximum flame luminescence.

Figure 5.16: Gas-phase normal $\langle u_z u_z \rangle_g$, radial $\langle u_z u_r \rangle_g$ and shear $\langle u_z u_r \rangle_g$ Reynolds stresses profiles at $z = 10, 15, 20, 30 \text{ mm}$ for case $\text{AcH}_{\text{III}}$. 
higher than those of case $H_{III}$. Also, a strong anisotropy is present. As the gas-phase evolves downstream, anisotropy persists in the center region where $n_d$ is relatively high (see Fig. 5.14).

**Droplet-size class properties**

To illustrate the characteristics of the droplets behavior, the mean axial $\langle U_z \rangle$ and radial $\langle U_r \rangle$ velocity of four droplet size-classes at three axial stations, $z = 10, 20$ and $30$ mm, are shown in Fig. 5.17. The class width was chosen as $10$ µm. All the points are an average of at least 200 samples. The mean velocity of the droplets in size class $0 - 6$ µm are considered to be flow tracers and thus represent the gas-phase velocity.

Figure 5.17: Mean axial $\langle U_z \rangle$ and radial $\langle U_r \rangle$ velocities profiles per size class for case $AcH_{III}$.

As in case $H_{III}$, an increase of the droplet slip velocity with the size-class is observed in the near atomizer region. The $\langle U_z \rangle$ downstream evolution shows that the slip velocity gradually decreases as a result of the momentum exchange between the two phases and, ultimately, the different size-class radial profiles exhibit a similar mean axial velocity profile at $z = 30$ mm. As expected for all droplets size classes the axial velocity component is larger than that of case $H_{III}$ (see Fig. 5.9).
Gas-phase temperature evolution in the downstream region

The gas-phase temperature profiles at \( z = 30, 40, 50 \) and 60 mm are shown in Fig. 5.18. On the left-hand side, the gas-phase mean temperature \( \langle T \rangle \) is presented along with the corresponding mean temperature gradients \((d\langle T \rangle/dr)\). On the right-hand side, the r.m.s. fluctuations \( T' \) are presented.

At \( z = 50 \) mm, \( r = 13 \) mm a sharp \( \langle T \rangle \) rise along with a substantial increase of \( T' \) is observed. Then, \( \langle T \rangle \) gradually decreases towards the centerline, however, remains significantly higher than the acetone ignition temperature \( (T_{ig} = 738 \) K). It can be concluded that, as in case \( \text{H}_{\text{III}} \), the mixture stoichiometry evolves from lean to richer mixtures from the most outer parts of the spray towards the center. The lean flammable mixture at the spray most outer parts engages into the oxidation process and the heat-release yields a rise in the gas-phase mean temperature. As one approaches the spray axis so as the vapor-fuel increases, the heat-release is gradually reduced and a decrease of \( \langle T \rangle \) takes place.

Fig. 5.18 shows additional information concerning the influence of air entrainment on the coflow inlet temperature profiles. It can be noticed that in the region between \( r = 20 \) and 40 mm, the coflow temperature remains nearly the same throughout the axial stations presented. This shows that up to \( z = 60 \) mm, the burner pipe diameter is wide enough to isolate the spray flame from the laboratory air entrainment and the flame is stabilized only by the hot-diluted coflow. Since no additional measurements were performed at higher axial stations, it is not possible to determine accurately where the laboratory air starts to perturb the acetone spray flame.

Gas-phase temperature histograms

The normalized temperature histograms of five radial positions are presented in Fig. 5.19 for case \( \text{Ach}_{\text{III}} \). The radial positions were selected to evaluate the trends in the temperature histograms along a plane located at \( z = 40, 50 \) and 60 mm. A bin size of 50 K was selected for all the histograms presented. The total number of samples in each histogram amounts to 1000. For each of the five radial positions selected, the corresponding mean \( \langle T \rangle \), root-mean-square temperature fluctuation \( T' \) as well as skewness \( (S) \) and kurtosis \( (K) \) of the temperature distribution are presented in the inset.

The axial station of \( z = 40 \) mm is located prior to the region where flame luminescence is visible. At \( r = 30 \) mm, \( \langle T \rangle \) and \( T' \) are 1368 K and 98 K respectively. This is a radial position in the hot-diluted coflow where no droplets are present. The temperature distribution is rather symmetric \( (S = 0) \) and
5.2 Results and discussion

Figure 5.18: Gas-phase mean temperatures \( \langle T \rangle \) (left) and r.m.s. fluctuations \( T' \) (right) at four axial stations. The dashed grey lines correspond to the corresponding mean temperature gradients \( (d\langle T\rangle/dr) \).
Figure 5.19: Normalized temperature histograms for five radial positions at $z = 40, 50$ and $60$ mm for case AcH III.
extends from 1100 K to about 1650 K. For all the radial position in the spray region, \( r = 0, 6, 12 \) and 16 mm the normalized temperature histograms shape are rather alike, yet, shifted to lower temperatures. From the normalized histograms in the spray region, it can be noted that no samples are present above 1650 K. This indicates that along the cross-section the gas-phase temperature decreases due to evaporative cooling and no ignition events take place in this region.

For the axial station of \( z = 50 \) mm, i.e. slight above the flame lift-off, some differences can be observed. From the coflow core to the radial position on the spray most outer parts, \( r = 15 \) mm, an increasing number of samples above 1650 K occurs. The histograms are shifted towards higher temperatures and spread over a larger range of temperatures as compared to \( z = 40 \) mm. An increase of \( \langle T \rangle \) is also observed (see inset). The increase of \( \langle T \rangle \) in the region wherein reaction occurs is contrary to what observed in some gaseous flames in hot-diluted coflow conditions studies. As shown by Oldenhof et al. [30, 31], peak temperatures in the cumulative density function of the temperature distributions are present, however, no augmentation of the mean temperature is observed.

An additional conclusion can be inferred from these observations. The oxidation process takes place at the spray most outer parts wherein the hot-diluted coflow mixes with the fuel vapor resulting from the droplets. In case H\(_{III}\), this is a region with low droplet number density, large \( d_{32} \) and where droplets below 10 \( \mu m \) are absent. The larger droplets penetrate into the hot coflow and leave a trail of fuel vapor behind, which depend on the convective and vaporization time scales. In case AcH\(_{III}\), as a result of the different atomization mechanisms, a substantial amount of droplets below 10 \( \mu m \) with higher initial velocities are present at the spray periphery. The time required for these droplets to respond to a change in the flow field velocity is small and, therefore, are quickly engulfed in the large scale mixing structures at the spray periphery and have sufficient time to vaporize and mix. Additionally, the acetone latent heat and boiling point is substantially smaller than that of ethanol, reducing the vaporization time scale by a factor of 0.7. Hence, the presence of small droplets in case AcH\(_{III}\) contributes to an earlier formation of an ignitable mixture at the spray periphery. However, one should note that the largest portion of the fuel is contained in the large droplets. A comparison of the convective time scale between cases H\(_{III}\) and AcH\(_{III}\) show an decrease by a factor of 2.9 for droplets of 15 \( \mu m \) and of about 2 for droplets of 45 \( \mu m \). This indicates that, although droplet vaporization occurs more rapidly in AcH\(_{III}\) as compared to case H\(_{III}\), the motion of the large droplets is considerably faster and the formation of an ignitable mixture takes place at a higher axial loc-
ation, this being in accordance with the finding that $A_{\text{CH}_{\text{III}}}$ lift-off height is higher than $H_{\text{III}}$.

5.3 Conclusions

A study was conducted in ethanol and acetone sprays issuing in identical hot-diluted coflow conditions. The bulk temperature and oxygen dilution were chosen to be representative of practical combustion systems operating in MILD conditions. The liquid mass flow rates were selected to yield sprays with identical Weber numbers. High-speed visualizations and complementary pointwise measurement techniques, such as LDA, PDA and CARS, were employed separately to unveil the atomization mechanisms as well as the spray flame structure and droplet dynamics. The current measurements complement other databases for modelling and numerical simulations of spray flames as well as liquid fuel atomization in hot-diluted conditions [43, 44].

High-speed visualizations revealed that the mechanisms involved on the atomization of the different liquid fuels are distinct. For ethanol spray flame, a liquid sheet exits from the atomizer nozzle and vaporization helps the local thinning of the liquid sheet and the subsequent disruption into ligaments and droplets. In the acetone case, cavitation takes place in the atomizer chamber. This originates a two-phase liquid-vapor flow issuing from the atomizer nozzle and changes drastically the initial gas-phase and droplet mean velocity as well as the local coflow conditions. Hence, a mathematical model of the two-phase liquid-gas flow that accounts for those effects is needed for numerical simulations.

The flame visual appearance differs greatly for both liquid fuels. For the ethanol spray flame case, the lift-off height observed is substantially lower than that of acetone which is contrary to what is predicted based on comparison of the enthalpy of vaporization. The measurement results show that the difference in lift-off height is a direct consequence of the vaporization inside the atomizer nozzle and, ultimately, the initial droplet momentum. Although acetone is a more volatile fuel, the large droplet convective time scales decrease more significantly and an ignitable mixture is originated at a higher axial location. Additionally, CARS measurements in the rich faint brush region downstream of the spray flame, show the occurrence of temperature samples above the fuel adiabatic temperature. This effect is attributed to the production of $H_2$ and show that detailed chemistry and preferential diffusion effects need to be account in the numerical simulations of the studied spray flames.
CHAPTER 6

Conclusions and recommendations

The results presented in Chapter 3, 4 and 5 have revealed many interesting phenomena relating to spray flames in air and hot-diluted coflow. An analysis of the results increases our understanding on the liquid breakup, flame structure and the stabilization processes in conventional and MILD conditions, which may be extended to combustion processes in advanced combustion systems. In this chapter, the main conclusions are summarized and the significance of these results is put in the overall context of MILD combustion. Suggestions are also presented in order to establish guidelines for other researchers willing to conduct further experimental studies.

6.1 Laboratory-scale burner design

A laboratory-scale burner was successfully designed and constructed. The burner consists of a pressure-swirl spray issuing in an coaxial flow of hot combustion products from a secondary burner. The secondary burner allows the use of seeding particles for laser-based diagnostics and offers several advantages as compared to other laboratory-scale burners in the literature used to emulate MILD conditions [29, 78] or the recirculation region of gas turbines [84]. In view of other researchers willing to reproduce the experiments, some conclusions of the current design and practical limitations are discussed below.

Laboratory air entrainment

The test section consists of a spray issuing in an open hot-diluted turbulent coflow. The open configuration was adopted to allow easy optical access for laser-based diagnostics. A disadvantage is that the velocity gradient between the hot-diluted coflow and the laboratory air gives rise to a growing shear layer starting at the edge of the outer pipe that perturbs the flame above a certain axial location. In this study, a outer pipe diameter of 160 mm was
selected to isolate the spray flame from the laboratory air for a large down-
stream distance. As compared to other laboratory-scale studies designed to
study MILD combustion processes \cite{19, 32}, the coflow diameter is double. The
ethanol spray flames cases (H\textsubscript{I}, H\textsubscript{II} and H\textsubscript{III}) are shielded from the laboratory
air up to \(z = 60\) mm. The acetone spray flame case (AcH\textsubscript{III}) has a smaller
spray cone. Since the droplets move mainly upwards and slowly disperse into
the hot-diluted coflow, the laboratory air influence is expected to be present
at a higher axial station. No measurements were performed above \(z = 60\) mm.
However, from the droplet spreading and the mean temperature profiles evol-
ution it is expected for the laboratory air to influence the flame at about
\(z = 90\) mm.

Care must be taken when comparing these results with those of other stud-
ies employing the same burner configuration. The use of other commercial
atomizers or liquid fuels with distinct physical properties from those used in
this study, may yield spray flames with different cone angles. In case of a
higher initial radial velocity component of the droplets, a displacement of the
outer flame-front towards larger radial positions will occur. Consequently, the
presence of laboratory air will be observed at lower axial positions. Other
researchers have produced alternatives that can slow down the growth of the
shear layer by, for example, enclosing the burner by a wind tunnel \cite{19}.

**Burner operational range**

The secondary burner design adopted in this study allows the use of seeding
particles for laser-based diagnostics while maintaining a temperature control
over a wide range, achieved by varying the fuel to air ratio, and preserving a
reasonably uniform field of burned gas velocity, temperature and composition
without experiencing any flame stabilization problems. These features fulfill
the needs for numerical simulations and model validation of turbulent com-
bustion models (see Section 2.1). Nonetheless, two limitations are associated
with the current burner design.

Firstly, the coflow temperature and oxygen dilution cannot be varied inde-
dependently unless air is diluted with \(\text{N}_2\) prior to the secondary burner. How-
ever, for the air mass flow rates selected, producing a coflow with lower oxygen
dilution requires experimental facilities beyond the budget available for this
study. Secondly, the establishment of lower oxygen dilution by reducing the
air-to-DNG ratio in the secondary burner leads to ‘combustion noise’. The
consequences are an intolerably high tonal noise and the unsteady behavior of
the spray flame.

For the experiments presented in this study, the effects of the length of the
outer pipe and the perforated plates positions were considered and optimized to eradicate this effect for a certain range of operating conditions. However, it shall be retained that, with the presence configuration, reducing the oxygen dilution of the coflow (and consequently increasing the coflow temperature) leads to the appearance of this effect. An increase of the air-to-DNG ratio suppresses this effect, however, it also increases of the characteristic flame-length in the secondary burner. Depending on the mass flow rates, the flames upon the secondary burner may interact with the bottom perforated plate deteriorating the homogeneity of the coflow. Hence, in order to study a specific range of coflow temperature and oxygen dilution, a compromise must be found between the outer pipe length, location of the perforated plates and the desired air-to-DNG ratio.

Relaminarization effects

The high temperatures result in partial relaminarization, through changes in the laminar viscosity and the subsequent decrease of turbulent energy at small scales. For the ethanol cases, above $z = 30$ mm the turbulence levels are considerably low (see Fig. 4.8).

Typically in industrial furnaces, the oxidiser stream that surrounds the fuel stream originates from strong coaxial hot air jets yielding higher turbulences levels than those observed in laboratory scale studies. As consequence, the partial relaminarization may result in much larger Damköhler numbers than those encountered in flameless combustion applications and low mixing rates in the far downstream region.

Thorough and extensive review of earlier research show that other burner configurations can overcome this limitation \[12, 99\]. In these studies, the burner consists of a ultrasonic atomizer used as the fuel injector and a turbulence-promoting plate in the burner. However, the liquid atomization and the specific-atomizer effects on the gas flow that, are of importance to practical combustion systems, are not represented.

6.2 Liquid breakup, spray flame structure and droplet dispersion in conventional and MILD conditions

Analysis of the results leads to the conclusion that, for ethanol sprays in hot-diluted coflow, the strong vaporization promotes the local thinning of the
Conclusions and recommendations

Liquid sheet and leads to an earlier fragmentation of the liquid jet than in air coflow. Nevertheless, the $d_{32}$ distribution in the near atomizer region remains similar in air and hot-diluted coflow conditions, i.e., small droplets at the center moving mainly upwards and increasing droplet size towards the spray edge. Depending on their initial momentum and Stokes numbers, droplets disperse into the coflow as the spray evolves downstream. The spray flame in air coflow is stabilized by flame propagation into a mixture of droplets-vapor-air and the flame propagation depends on the local gas flow conditions, droplet size, droplet density and the self-acceleration of the chemical reaction rate. In the hot-diluted coflow, the gas flow already contains the energy needed to vaporize the liquid droplets and self-ignite. Hence, small droplets are quickly vaporized and the combustion process is mainly dependent on the mixing of the fuel vapor with the entrained hot-diluted coflow and the ignition delay time.

Higher coflow temperatures enhance the droplet vaporization rates and the chemical reaction rate and, therefore, promote the appearance of an earlier ignitable mixture. Consequently, two phenomena are observed: 1) reduction of the flame lift-off height for the same injection pressure (for nearly identical droplet convective timescale), 2) earlier formation of intermediate species such as $\text{H}_2$ that rapidly oxidize and leads to an increase of peak temperatures at lower axial stations in the outer flame-front.

From the comparison of the acetone and ethanol spray flames it can be concluded that the liquid fuel properties have a potential impact in the atomization mechanisms and the resulting gas flow field in the near-atomizer region which, ultimately, yield substantial differences in the droplet distribution and dispersion far downstream. Consideration of the aforementioned points is especially important for the flame structure. Also, the temperature peaks at the outer flame-front suggests that both the entrained fluid temperature and the fuel chemical kinetics play a key role in achieving MILD conditions.

6.3 Recommendations

Potential for planar imaging applications

In this experimental study, complementary pointwise laser diagnostics techniques and high-speed visualizations were employed separately. Although the experimental database provides a building-block for the improvement of combustion models in turbulent multiphase reacting flows, the application of other laser-based diagnostics are likely to yield major scientific breakthroughs.

The gas flow field was determined from the smaller droplets measured
by the PDA. However, this procedure is limited by the availability of small droplets and the gas flow field only was partly unveiled. In order to retrieve complete information on the gas-phase velocity field, the liquid fuel could be, for example, doped with Rhodamine and Particle Image Velocimetry (PIV) employed. The emitted fluorescence from the droplets is shifted from the Mie scattering of the seeding particles and, therefore, it is possible to distinguish the droplets from the seeding particles in the PIV images by means of dichroic filters in the CCD cameras [21]. Furthermore, simultaneous OH Planar Laser-Induced Fluorescence (OH PLIF) could give qualitative information of the structure and dynamics of the inner flame-front and the stabilization zone.

Value for model validation

The experimental database obtained in this study provides sufficient information to define the boundary conditions of the dilute regions in the sprays. The initial boundary conditions of the dilute-phase can be determined either by 1) using phenomenological model of spray breakup or 2) by the measured droplet and gas-phase statistical properties at the start of the dilute-region. For the first approach, the input parameters, i.e. the liquid fuel mass flow rate, injection pressure and temperature prior to the atomization were measured and, along with the high-speed visualizations, gives rise to opportunities for the development and validation of new breakup models accounting for vaporization. For the second approach, a comprehensive dataset is available at several positions in the near atomizer region of each test case.

The measured gas-phase statistics at the atomizer exit plane location revealed that the presence of the spray leads to an acceleration of the coflow velocity adjacent to the spray. To the author knowledge, this phenomena was never addressed before and future experimental studies shall strive to measure adequately the coflow inlet conditions so that input boundary conditions for numerical simulations are properly described.

It would be of interest to use these results for validation of LES and RANS methods. The database reported here contains several cases qualitatively different from each other making it possible to test range of validity of models. The cases of air coflow and hot-diluted coflow are shown to lead to drastically different flame structure. The results indicate changes in local flame type and strength of heat release that a priori seem difficult to predict by models using only a single type of local flame structure or only simple chemistry. The database also offers the opportunity to test models for their ability to cover different fuel composition and chemical mechanisms.


[63] M. Linne. Imaging in the optically dense regions of a spray: A review of


Acronyms

**HiTAC**  High Temperature Air Combustion  
**EEC**  Excess Enthalpy Combustion  
**FLOX**  Flameless Oxidation  
**MILD** Moderate and Intense Low-oxygen Dilution  
**PDA**  Phase Doppler Anemometry  
**LDA**  Laser Doppler Anemometry  
**PIV**  Particle Image Velocimetry  
**CARS** Coherent Anti-Stokes Raman Spectroscopy  
**DNG**  Dutch Natural Gas  
**NA** Nonreacting spray in air coflow - I  
**A** Spray in Air Coflow - II  
**H** Spray in Hot-diluted coflow - I  
**H** Spray in Hot-diluted coflow - II  
**H** Spray in Hot-diluted coflow - III  
*d* Sauter mean diameter  
**AcH** Acetone Spray in Hot-diluted coflow - III
Acknowledgements

I am deeply grateful to my promotor Prof. Dirk Roekaerts who has served as a valuable asset in producing solid scientific ideas and a source of a never-ending kind. The guidance and support he has provided, his integrity and character have made a deep impression upon me. It has been a great pleasure and thank you for all moments we shared.

I am also grateful to Eric van Veen for the generous assistance during the experimental campaigns. He is one of the most intelligent, honest and reliable individuals that I have ever met. Thank you for the conversations we had about the CARS setup and for handing the measurements with high level of professionalism.

I also would like to thank Prof. Mark Tummers the guidance and support to write the articles. I will always be impressed by his persistence in sharpening arguments and his strive to see no stones unturned. Together with Dirk Roekaerts and Eric van Veen, this unit taught me the great value of communication and scientific research.

Thanks also to my closest colleague Milos Bilvarske, with whom I have spent many hours in the laboratory and whose strong support in the hectic period of writing the thesis is unforgettable. Also, I was fortunate to work in close collaboration with Louis Armen, Ernst Oldenhof, Gerasimos Sarras, Likun Ma and Michael Stöllinger which shared with me views and perspectives as modelers of turbulent combustion and with whom I had enlightening scientific discussions. I also want to pass on my warmest regards to all colleagues of mine at the multi-scale physics and process and energy department and fluid mechanics laboratory such as Carsten Trapp, Javier Fernandez, George Krintiras, Nafiseh, Pedro Costa, Daniele Fiscaletti and Valentina Valori. This has been a bright period of life thanks to all of you.

Last, but not least, thanks to Elsa Rodrigues, which always provided me the emotional support I needed in the most difficult times.
About the Author

Hugo Ricardo Correia Rodrigues
Born on October 26th, 1984
in Lamego, Portugal

1997 - 2002 High school,
Licéu Latino Coelho, Lamego, Portugal

2002 - 2008 Bachelor and Master of Science in Mechanical Engineering,
Faculty of Engineering of the University of Porto, Portugal

2008 - 2009 Research Master in Environmental Fluid Dynamics,
von Karman Institute for Fluid Dynamics, Belgium

PhD Researcher,

2009 - 2013 Department of Multi-Scale Physics,
Faculty of Applied Sciences, Delft University of Technology,
The Netherlands

2013 - 2014 Laboratory for Aero- and Hydrodynamics,
Faculty of Mechanical, Maritime and Material Sciences (3mE),
Delft University of Technology, The Netherlands

2014 - 2015 Senior Research Engineer,
Luzern University of Applied Sciences and Arts,
Bioenergy Research Laboratory, Switzerland
2015 - now  **Research & Development Engineer**,  
European Organization for Nuclear Research (CERN),  
Accelerator Technology Department, Cryogenics Group,  
Geneva, Switzerland
Journal Publications

Conference proceedings


Abstracts of presentations at conferences


Abstracts of poster presentations


