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Highlights

- We visualized flash-boiling jets at an unprecedented frame rate of 5 Million frames per second. At such high frame rate we observed for the first time the evolution of the bubble expansion and the burst mechanisms, responsible for the jet atomization.
- For developed flash-boiling conditions, minimum pixel intensity profiles revealed that droplets are ejected in all directions from the nozzle. This crucial fact was translated into spray angles larger than 300° .
- The detailed close up to the nozzle revealed that the droplet size distribution of the spray is wide, contrary to the common belief. Using a simple argument we infer that the smallest droplets are on the hundreds of nanometer range. This means that the drop size distribution extends through almost four orders of magnitude.
- We measured velocities up to 140 m/s during the expansion of bubbles from the core of the jet. Such velocity is a bit less than half of the speed of a 0.22 bullet, an unprecedented velocity for expanding bubbles.

Ultra-high speed visualization of a flash-boiling jet in a low-pressure environment

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Abstract

We visualize the flash-boiling atomization of liquid jets released into a low-pressure environment at frame rates of up to five million frames per second using a long distance microscope. Such temporal resolution allowed us to capture the details of the bubble expansion mechanism, responsible for the jet atomization, for the first time. We document an abrupt transition from a laminar to a fully external flashing jet by systematically reducing the ambient pressure. We perform experiments with different volatile liquids, ejected through micro-nozzles with different inner diameters. Surprisingly, minimum pixel intensity projections revealed spray angles close to $\theta_s \sim 360^\circ$ and speeds of bubble expansion up to 140 m/s. Particle tracking shows that ejected droplets achieve speeds much larger than the jet velocity and drop sizes order of magnitude smaller than the diameter of the nozzle. Furthermore, hole growth speeds measured on the bubbles film in combination with Taylor-Culick predictions suggests that the smallest droplet sizes are on the hundreds of nanometer or submicron range, which contravenes the general belief that flash-boiling atomization results in uniform drop sizes.

Keywords: Flash-boiling jet, Ultra-high speed, Jet breakup.

2010 MSC: 00-01, 99-00

1. Introduction

Leaks of compressed fuel such as liquefied petroleum gas (LPG) or natural gas (LNG) involve a great risk of explosion. In 2016 there were 306 incidents with gas pipelines according to the Pipeline and Hazardous Materials Safety Administration (PHMSA). These incidents resulted in 16 fatalities, 82 injuries and more than 300 million dollars in related costs, only in the USA¹. However, not every fuel leak entails the same risk. In fact, small fuel leaks from pressurized containers or pipes are fairly common. In most cases the leaks are small

and the fuel escapes as pure vapor. In this situation, evaporation is fast compared to the flow rate of the leak, allowing detection and correction. However, the risk increases disproportionately when the flow rate of a leak is high. At a high flow rate, the fuel reaches the atmosphere as superheated liquid. Then, an explosive atomization and vaporization occurs, usually referred as flash-boiling. The fine spray of droplets formed in this way can reach very far places in unexpected directions, even far from the fuel source. This phenomena could result in the formation of large explosive clouds whose location is difficult to predict (Bjerketvedt et al., 1997). Out of control, flash-boiling could thereby have devastating consequences. On the other hand, the fine sprays produced with flash-boiling could be desirable in a controlled environment. For example, for producing a very fine spray of droplets of a uniform size distribution that improve combustion efficiency (Sher et al., 2008). This is desirable in some applications such as fuel injectors (Chen et al., 2013;

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¹Information available online from the official website of the PHMSA on <https://hip.phmsa.dot.gov/analyticsSOAP/saw.dll?Portalpages>

Vetrano et al., 2013; Lamanna et al., 2014; Bohon et al., 2015; Ju et al., 2015), but also necessary when driving pressures cannot exceed certain limit (Sher et al., 2008; Zeigerson-Katz and Sher, 1998). Flash-boiling atomization usually coexists simultaneously with other mechanisms of spray formation, such as turbulence, bubbles within the flow and coaxial gas flows. These external influences had made the study challenging and clouded its understanding. In the following paragraphs we examine some of these atomization mechanisms.

1.1. Mechanical effects on the spray formation

Liquid jets injected into the atmosphere through nozzles of circular cross section exhibit distinctive drop sizes that vary with the jet diameter as well as the exit velocity of the flow. At very small speeds, the drop breakup occurs due to the most unstable mode of the Rayleigh-Plateau instability (de Gennes et al., 2013). In this scenario, the wave number correlates to the diameter of the liquid jet as $\kappa^{-1} \approx 0.717 D$ (Chandrasekhar, 2013). Drops formed in this way have sizes similar to the inner diameter of the nozzle. At higher speeds, the jet breaks into slightly smaller drops because short wavelength disturbances at the interphase are increasingly the most unstable. This breakup regime is known in the literature as the First Wind (Reitz and Bracco, 1982; Lin and Reitz, 1998). At very high speeds, the drop diameter becomes much smaller than the nozzle's size. Two main regimes exhibit these small sized droplets, the Second Wind and the atomization regime (Lin and Reitz, 1998). For nozzles of circular cross section, these last two regimes are only achieved by applying very large driving pressures (Sher et al., 2008). Using such large pressures guarantees a large Reynolds (Re) and Weber (We) numbers. For different applications, these numbers are either calculated with the properties of the liquid or with the gaseous phase around the jet (Ashgriz, 2011). Large Reynolds numbers imply that either the liquid jet or its surroundings are turbulent. A large Weber number indicates that the surface of the liquid jet is highly unstable, either due to the interaction of the free surface with the liquid or with the surrounding gas. The transition between regimes due to the interaction of these parameters were discussed in the review of Lin and Reitz (1998).

For applications where such large driving pressures are not available, specially designed nozzles have been developed to disrupt and atomize the jet. Such nozzles are optimized to achieve uniform drop sizes and control the spray angles. On experimental studies, nozzles with annular cross section have been designed with an inner tube carrying either gas (Lasheras et al., 1998; Lee et al., 2014; Charalampous et al., 2009; Wahono et al., 2008; Duke et al., 2015) or liquid (Sivakumar and Kulkarni, 2011). These nozzles promote the atomization by forming jets of annular cross section that are unstable due to capillary effects or promote the breakup through Kelvin-Helmholtz instability driven by the velocity difference across the interphases. Disrupting the jet with mechanical forces usually requires large driving pressures. In situations where applying large driving pressures is not possible, thermal mechanisms have been quite successful in producing fine sprays. Regarding spray formation, mechanical forces are insignificant when compared with strong thermodynamical driving mechanisms, such as flash-boiling. We discuss this mechanism below.

1.2. Flash-boiling thermodynamics and parameters

Flash-boiling dynamics has been applied to increase the spray angles of jets, reduce their mean droplet size and reduce driving pressures (Ju et al., 2015). The main ingredient of a flash-boiling spray is a superheated liquid, phenomenon occurring when the liquid's temperature T_l rises above its boiling temperature T_b at the ambient pressure (Polanco et al., 2010). The degree of superheating of a liquid is quantified with the temperature difference $\Delta T = T_l - T_b$. Conceptually, the easiest way to superheat a liquid is by increasing its temperature T_l . Alternatively, decreasing the ambient pressure reduces the boiling temperature as $T_b = T_b(P_\infty)$, also resulting in superheating. Then, the degree of superheating is a function the pressure as well as the temperature. The onset for boiling conditions is given by an equilibrium between the boiling temperature and the vapor pressure of the liquid P_v . A common way to relate these variables is with the Clausius-Clapeyron equation, usually expressed as $P_v = P_o \exp(-h_{fg}/RT_l)$, where P_o is a constant, h_{fg} is the enthalpy of vaporization and R is the universal gas constant. Fitting the experimental data from Stiles and Cady (1952), Dunlap et al. (1958), Crowder et al. (1967) and Dias et al. (2005), we

estimated that $P_o = 10.35$ GPa and $h_{fg} = 31.7$ kJ/mol for perfluorohexane (PFnH), one of the liquids used in this investigation. Alternatively, the vapor pressure of other liquids have been estimated using the Antoine equation, which relates the vapor pressure of the liquid with its temperature as $P_v = 10^{[a-b/(c+T_i)]}$. The constants a , b and c are given in Table 1 for the working fluids used in this investigation, except for PFnH. These values are used to estimate the boiling temperature when the ambient pressure has been reduced.

Table 1: The Antoine coefficients of the liquids used in this investigation. MeOH, EtOH and 1-PB stand for methanol, ethanol and 1-bromopropane.

Liquid	a	b	c	T_{min} [°C]	T_{max} [°C]
MeOH	7.87863	1473.11	230	-	-
EtOH	8.20417	1642.89	230	-57	80
1-PB	7.03766	1259.836	232	-53	71

If the temperature of the liquid is kept constant while varying the ambient pressure, the ratio between the ambient pressure and the vapor pressure P_∞/P_v is the leading dimensionless parameter characterizing the deviations from thermodynamic equilibrium (Lamanna et al., 2014; Luo and Haidn, 2016). This pressure ratio is a key element of the chemical potential ($\Delta\mu \sim k_B T \ln[P_v/P_\infty]$) and it is also correlated to the rate at which vapor nuclei form (Lamanna et al., 2014; Luo and Haidn, 2016). It has commonly been assumed that flash-boiling atomization is mainly dominated by bubble nucleation mechanisms (Park and Lee, 1994; Lamanna et al., 2014). While a superheated liquid is susceptible to form bubble nuclei, the bubble growth rates within the jet are proportional to the modified Jacob number

$$Ja = \frac{C_{pl}\Delta T}{h_{fg}} \frac{\rho_l}{\rho_v}, \quad (1)$$

where C_{pl} is the specific heat capacity of the liquid. Values of the specific heat capacity and the enthalpy of vaporization for the working fluids in this investigation are presented in Table. 2. The densities of the liquid and the vapor are ρ_l and ρ_v , respectively. The vapor densities shown in Table 2 were evaluated at ambient pressure and at injection temperature (Kitamura et al., 1986), using the ideal gas equation ($P_\infty/\rho_v = RT_i/M$), where M is the molar mass in kg/mol. The Jacob number establishes the limits for a phase change to occur and

therefore it has been used to estimate the onset of flash-boiling (Vetrano et al., 2013).

Table 2: Thermal properties of the working liquids

Liquid	C_{pl} [J/mol·K]	h_{fg} [kJ/mol]	ρ_l [kg/m³]	ρ_v [kg/m³]
PFnH	248	31.7	1700	13.96
MeOH	79.5	38.7	790	1.32
EtOH	112.4	42.5	786	1.90
1-PB	134.6	31.8	1354	5.1

1.3. Connection between the mechanical and thermodynamic effects

Flash-boiling atomization results from a combination of thermodynamical and mechanical effects. On a jet with a fixed Jacob number, a liquid with an agitated interphase has a higher flashing potential than an unperturbed surface. A relationship between the vapor Weber number and the critical Jacob number marks such a boundary for flash-boiling (Kitamura et al., 1986),

$$Ja \phi = k We_v^{-1/7}. \quad (2)$$

The vapor Weber number in Eq. 2 is defined as $We_v = \rho_v v^2 d / \sigma$, where d is the inner diameter of the nozzle, v is the discharge velocity of the jet and σ is the surface tension. The parameter ϕ was derived experimentally taking into account the bubble nucleation growth rate (Kitamura et al., 1986). This parameter is calculated as $\phi = 1 - \exp[2300(\rho_v/\rho_L)]$. The prefactor k has two values according to the different flashing regimes. Macroscopic bubbles start altering the evolution of the spray when $k \geq 55$, while a fully flashing jet develops when $k \geq 150$ (Lamanna et al., 2014). These two boundaries between the flashing regimes are shown as the solid and broken lines in Fig. 1. The relationship from Eq. 2 has been demonstrated experimentally and the prefactors have been determined empirically (Vetrano et al., 2013; Lamanna et al., 2014). The parameter ϕ from Eq. 2 is $\phi \approx 1$ for PFnH, methanol (MeOH) and ethanol (EtOH). For 1-bromopropane (1-PB) the value of the parameter is approximately $\phi \approx 0.6$. In principle, jets with Weber numbers below the threshold established by Eq. 2 will not flash. The small exponent in Eq. 2 implies that changes in the thermal conditions are far more important than a turbulent interphase. So, despite the mechanical and thermal effects are correlated,

the dependance of the Ja on the We_v is weak. In Fig. 1 we have added symbols corresponding to the experimental conditions of the Figs. 3-8 and 10.

Similar physics has been reported for evaporating liquid sheets. In this investigation, Pavlenko et al. (2009) found that the dimensionless distance between jets (resulting from the breakup of the film) remains constant with the Reynolds number for a stationary heat release. However, an agitated interphase plays an important role in the formation of these patterns. Later, Pavlenko et al. (2014) demonstrated that small disturbances on the surface of liquid layers result from instabilities on the wave fronts.

Figure 1: Parametric space relating the vapor Weber (We_v) and the Jacob (Ja) numbers. The experimental conditions of different Figs. 3-8 and 10 are shown with symbols.

1.4. Experimental studies and measurements

Flash-boiling atomization has been characterized experimentally in numerous studies by investigating the effect of several parameters and conditions. To assess the effect of the ambient pressure, experiments have been performed in vacuum (Kurschat et al., 1992; Lecourt et al., 2009; Pavlenko et al., 2010; Vetrano et al., 2013; Du et al., 2013; Xu et al., 2013; Lamanna et al., 2014; Luo and Haidn, 2016; Guo et al., 2018) as well as in high pressure chambers (Zhang et al., 2015; Cai et al., 2017). Water has been commonly used as a test liquid (Park and Lee, 1994; Du et al., 2013; Günther and Wirth, 2013; Pavlenko et al., 2013; Cai et al., 2017; Sinha et al., 2018).

Flash-boiling water has been used to increase the relative humidity (Cai et al., 2017). Other experimental investigations have used common organic solvents such as acetone (Lamanna et al., 2014), EtOH (Vetrano et al., 2013; Lamanna et al., 2014; Lecourt et al., 2009; Xu et al., 2013) and MeOH (Xu et al., 2013; Zhang et al., 2015; Thompson and Heister, 2016). Also, non-common organic solvents such as Dimethyl ether (DME), an isomer of EtOH, have been tested (Ju et al., 2016a). Due to their high volatility, refrigerants such as liquid nitrogen (Luo and Haidn, 2016), Freon-11 (Pavlenko et al., 2010), PFnH (Kurschat et al., 1992), R134a (Ju et al., 2015; Zhifu et al., 2012; Ju et al., 2016b; Wang et al., 2017) have been investigated as well. Organic compounds with high vapor pressures can be included in this category, such as Acetaldehyde (Lecourt et al., 2009). Fuels like gasoline are very relevant for the automotive industry (Xu et al., 2013; Zeng et al., 2012; Guo et al., 2017) as well as rocket propellants, such as monomethylhydrazine (MMH) (Lecourt et al., 2009) and nitrogen tetroxide (NTO) (Lecourt et al., 2009), for the aeronautical industry. These studies also cover flash-boiling atomization in liquid mixtures, such as Chlorodifluoromethane (CHClF₂ or R-22), and silicon oil (Levy et al., 2016). In addition to these studies, it has been proposed that the mechanism of phase change of liquids, exhibiting retrograde condensation, is different (Vieiras and Simoes-Moreira, 2007). This has led to flash-boiling experiments focussed on retrograde liquids such as isooctane (Lamanna et al., 2014; Vieiras and Simoes-Moreira, 2007). The above experimental studies have injected the liquids using nozzles of different sizes ranging from 900 μm (Günther and Wirth, 2013), down to 150 μm (Vetrano et al., 2013; Lamanna et al., 2014).

Flash-boiling sprays are characterized through the measurement of various physical parameters. Some of these are the spray angle θ_s , the size distribution and the velocity of ejected drops. Spray angles have been measured and reported for different liquids. Some studies have reported spray angles smaller than 90° (Ju et al., 2015, 2016b,a; Guo et al., 2017). These studies were performed on very different conditions and even using different liquids, such as R134a, gasoline and DME. However, angles larger than 90° were reported with water (Du et al., 2013; Park and Lee, 1994) as well as R134a (Zhifu et al., 2012). Luo

and Haidn (2016) measured a maximum angle of 120° using liquid nitrogen. Performing experiments with different liquids, Lamanna et al. (2014) found that acetone can reach a spray angle of 160°, a value marginally larger than when using EtOH and isooctane. Finally, the largest angle reported was 180° with Freon-11 (Pavlenko et al., 2010). Even if the definition is not entirely the same in these studies, there is a considerable difference in the values reported. One reason might be that these angles have been reported using different imaging methods. Some of them used high-speed photography (Du et al., 2013; Ju et al., 2015; Lamanna et al., 2014; Pavlenko et al., 2013; Vetrano et al., 2013; Zhifu et al., 2012; Zhang et al., 2015; Guo et al., 2017; Ju et al., 2016b,a; Thompson and Heister, 2016; Wang et al., 2017), while others took photographs with short light pulses or short exposure times (Park and Lee, 1994; Kurschat et al., 1992; Guo et al., 2018). Alternatively, pulsed lasers have been used to illuminate since they emit light during very short times (Xu et al., 2013; Lecourt et al., 2009; Günther and Wirth, 2013; Sinha et al., 2018). In some cases, the pulsed lasers are used as part of a velocimetry technique such as PIV.

Recordings of the flash-boiling atomization are usually carried out in the dilute region of the spray (Lamanna et al., 2014). In this region the spray has achieved thermodynamical equilibrium and the breakup is dominated by mechanical effects (Polanco et al., 2010; Lamanna et al., 2014). There is a general agreement that flash-boiling results from bubbles nucleating and expanding, disrupting the liquid jet into droplets (Polanco et al., 2010; Sher et al., 2008). However, the temporal evolution of the expansion of these bubbles has never been observed (Lamanna et al., 2014). For experiments recorded with high-speed cameras, either the field of view is much wider than the nozzle or the exposure time is long compared with their expansion speed. Meanwhile, in experiments recorded with light short pulses or pulsed lasers, the time-evolution has been lost between frames. Therefore, in previous experimental realizations the details of the explosive phase transition have not been observed in detail. There are two main reasons for this gap. The first of them is that the region is very narrow, making it difficult to close up imaging. The second is that this is the region where the fastest phenomena occur. However, this region is of great interest because it is where

the non-equilibrium thermodynamic effects form the spray. Acquiring information where thermodynamic changes occur might help developing a general criteria to predict the behavior of flash-boiling jets, now still missing (Polanco et al., 2010).

In this paper we study experimentally the bubble expansion mechanism resulting from the explosive phase transition. For the first time, we visualized the flash-boiling atomization of different liquids very close to the nozzle using a long distance microscope objective. To capture the temporal evolution of bubble expansion, we recorded using an ultra-high speed video camera at up to 5 million frames per second (fps). The details of our experimental setup are described in the following section.

2. Experimental setup and methods

A sketch of the experimental setup is presented in Fig. 2(a) and it consists on a micro-nozzle injecting a liquid jet within a vacuum chamber, a ultra-high speed camera and a laser illuminating from the back. A vacuum gauge measures the pressure within the chamber. The temperature of the liquid is measured with a thermocouple just before the discharge. The driving pressure is controlled with a pressure regulator connected to a nitrogen tank. The liquid is kept at constant temperature. The level of the liquid is kept constant with a long tubing that remains elevated around one meter above the nozzle. Each of these elements is further described below.

2.1. Low pressure chamber

To create a stable low pressure environment a custom vacuum chamber was designed and built. The chamber had four acrylic windows, one on the front, one on the back and two at each side of the nozzle. A vacuum pump Adixen Pascal 2021SD was used to lower the pressure within the chamber. We connected the vacuum pump with the chamber using vacuum joints. The pressure within the chamber was measured with a digital pressure gauge Wika CPG1000. With this system we achieved a minimum reproducible pressure of 10^{-3} kPa within the vacuum chamber.

2.2. Hydraulic system driving the jet

The working fluids were injected at different driving pressures. The driving pressure was supplied

Figure 2: (a) Sketch of the experimental setup. Liquid jets of PFnH are discharged through nozzles of glass (b-e) and metal (f,g) within a vacuum chamber and recorded with a ultra-high speed camera. The d of the nozzles increases from left to right, being 30(b), 45(c), 83(d), 98(e), 100(f) and 150 μm (g). Videos of cases (e) and (f) are included as additional material.

by a nitrogen line available in the lab, that could reach up to 8 bar. To achieve higher pressures, we connected a nitrogen tank that allowed us to increase the maximum driving pressure up to 12 bar. The outlet pressure of either the line or the tank was controlled with pressure regulator (Iwashita Instruments AD3000C). The temperature inside the laboratory was controlled at 22°C. A thermo circulator was used to raise and keep constant the temperature of the liquids. The thermo circulator is a Phoenix II from Fischer Scientific and it is capable of raising the temperature of water up to 100°C. The temperature of the liquid was kept constant while the pressure regulator was connected to a long pipe. The pipe ascended from the pressure regulator up to one meter above the vacuum chamber. The highest part of the pipe was connected to a valve where the liquid was filled. At the same height of the nozzle the pipe had a transparent section within, to verify the amount of remaining liquid. This allowed us to keep the line filled with liquid, maintaining a constant volume and avoiding air bubbles within the flow. The pipe filled with liquid was connected to a stopcock valve. A custom fit was manufactured to connect the nozzle with the stopcock valve. The custom fit had a thread that was tightly screwed up to the bottom of the vacuum chamber. When temperatures higher than the ambient were required, the thermo circulator radiated hot liquid around the pipes carrying the working liquid. The liquid within the thermo circulator flows in the opposite direction of the working liquid,

forming a counterflow heat exchanger. Due to the possible heat losses in the pipe, the temperature was measured immediately before entering the vacuum chamber. This measurement was performed using a thermocouple Omega HH806AU, depicted in Fig. 2(a). Nozzles were tightly attached to the custom fit within the vacuum chamber. Previously calibrated, the driving pressure was selected to produce liquid jets with unperturbed interphases. The valve below the nozzle, immediately before the vacuum chamber, was opened shortly before recording the jet. This produced fresh liquid coming into the chamber, avoiding internal flashing. Internal flashing refers to flash-boiling before the discharge from the nozzle tip.

2.3. Ultra-high speed video recordings

The flash-boiling experiments were visualized using a ultra-high speed video camera, Kirana from Specialized Imaging. The camera records 180 frames at up to 5 million frames per second with a resolution of 924×768 pix. The long working distance objective on the camera was a Leica Z16 APO. The experiment was illuminated with a laser illumination system SI-LUX 640 and controlled with a laser safety unit, both from Specialized Imaging. This system consists on 180 diode lasers, each corresponding to every frame. The camera and the microscope objective are depicted in Fig. 2(a) and sample images of the free jets are seen in Fig. 2(b-f). The camera together with the long distance microscope lens were placed next

to the front window. The laser system and the backlight illumination resulted in shadow images with good contrast.

2.4. Working fluids

Our main working fluid was Flutec-PP1, whose chemical composition is C_6F_{14} or perfluoro-n-hexane (PFnH). However this liquid is described in the commercial MSDS as a PFnH isomer, perfluoro-2-methylpentane. Unfortunately, its exact chemical formula is not given in its MSDS, which does not allow to be sure of its thermodynamic properties. For practical purposes, we described the thermodynamic properties of Flutec-PP1 as pure PFnH. Other volatile liquids capable of flash-boiling at our working pressures were used, such as methanol (MeOH), ethanol (EtOH) and 1-bromopropane (1-PB). The physical properties of these liquids at 22°C and one atmosphere are shown in Table 3.

Table 3: Physical properties of the working liquids at 22°C. The temperature in α and β is 25 and 20°C, respectively.

Liquid	μ [mPa·s]	σ [mN/m]	P_v [kPa]	T_b [°C]
PFnH	0.7 ⁰	11.5	25	57.1
MeOH	0.59	22.5	19	64.7
EtOH	1.13	22.1	6.62	78.4
1-PB	5.24 ^{β}	25.9 ^{β}	19.5 ^{β}	71

The values of the vapor pressure in Table 3 indicate that PFnH is the most volatile of the liquids. Besides, the surface tension of PFnH is the lowest of these liquids. Fig. 2(b-g) shows jets of PFnH at atmospheric pressure, but from nozzles with inner different inner diameters. In our experiments, we calibrated the driving pressure with the velocity of the jet, allowing us to create jets whose surface is smooth. Besides the properties of the working liquids, the inner diameter d of the nozzle plays an important role determining the vapor Weber number. In the following section, we describe the nozzles employed in this investigation.

2.5. Metal and glass nozzles

Liquid jets were discharged using metal and glass nozzles. The metal nozzles were flat dispensing tips (Precision Tips from Nordson) with an inner diameter (d) of 100, 150 and 200 μm . These tips were fabricated of stainless steel. Nozzles with

Table 4: Summary of the dimensionless numbers.

Liquid	d [μm]	Ja	We_v	P_∞/P_v [$10^{-3}, 4$]
PFnH	30	[-33, 98]	[0.14, 0.6]	[$10^{-3}, 4$]
	45	[-33, 98]	[22, 32]	[$10^{-3}, 4$]
	68	[-33, 98]	[1.3, 44]	[$10^{-3}, 4$]
	83	[-33, 98]	[0.14, 36]	[$10^{-3}, 4$]
	98	[-33, 98]	[27, 69]	[$10^{-3}, 4$]
	100	[-33, 98]	3.6	[$10^{-3}, 4$]
MeOH	135	[14, 41]	[0.6, 10]	[1, 4] 10^{-1}
	150	[28, 41]	[1, 10]	[1, 2] 10^{-1}
	200	[-50, 95]	[1.2, 6.6]	[$2 \times 10^{-3}, 5$]
	300	[9.2, 41]	[1, 5.1]	[1, 5] 10^{-1}
EtOH	66	36	[0.41, 5]	10^{-1}
	100	[-61, 100]	[0.6, 22] 10^{-3}	[$2 \times 10^{-4}, 15$]
	140	36	[0.4, 5]	10^{-1}
	169	27	[0.4, 5]	2×10^{-1}
	300	[0, 27]	[0.4, 5]	[$2 \times 10^{-1}, 1$]
	480	[6.6, 8.8]	[1, 9.8]	[6, 7] $\times 10^{-1}$
1-PB	200	[-54, 112]	[2.8, 5.7]	[$5 \times 10^{-4}, 5$]

smaller inner diameters were fabricated *in situ* from borosilicate glass tubes using a micro-puller Model P-1000, from Sutter Instruments. Initially, the glass tubes had an outer diameter of 1 mm and 10 cm of length. The range of inner diameters obtained with this technique varied from 30 to 480 μm . Examples of PFnH jets produced by these nozzles are shown in Fig. 2(b-g). In these images, the inner diameter of the nozzles is increasing from left to right. Jets produced by nozzles made of glass can be observed in Fig. 2(b-e) and metal nozzles are seen in Fig. 2(f,g). In Fig. 2(b,e) we observe that far from the nozzle the jet breaks into droplets due to the Rayleigh-Plateau instability. All of these nozzles had a circular cross section that resulted in stable cylindrical jets when ejected at a low driving pressures and normal ambient atmospheric pressure, as seen in Fig. 2(b-g).

2.6. Dimensionless parameters explored

The combination of thermodynamical and mechanical parameters can be summarized on Table 4. The table includes the working liquids, the inner diameters of the nozzles d as well as ranges for the Jacob (Ja), vapor Weber (We_v) numbers and pressure ratios (P_∞/P_v).

2.7. Image processing and analysis

The ultra-high speed camera acquires 180 frames per recording, regardless of the frame rate. Some frames were ignored during the image processing due to the lack of light in the image or ghosting. The images were pre-processed by balancing the light intensity along the video recording. The background was calculated with the following steps. First, we calculated the maximum pixel intensity projection during the length of the video recording. The nozzle was still visible in the resulting image. Then, we masked the region around the nozzle to create an uniform background. Dividing the video recording by the background, enhanced the quality of the image. This process was performed using ImageJ (Rueden et al., 2017), an open source software. The image processing allowed us to visualize the flash-boiling atomization in great detail.

To calculate the spray angle, we computed the minimum pixel intensity projection (MPIP) from the processed video recording. This technique computes the minimum value of each pixel of the recording. The MPIP shows the trajectories of dark objects that crossed through the field of view, making the technique ideal to visualize the direction of drops ejected from a spray.

The MPIP also reveals the probability of finding a drop in a determined trajectory. To quantify the distribution of the path lines along the angle, we computed the radial average pixel intensity at different angles. First we inverted the image and then computed the average pixel intensity across lines of the same length.

3. Results

3.1. Phenomenological description

3.1.1. Axisymmetry close to the nozzle

When the ambient pressure is reduced slightly below the vapor pressure of the liquid, there is usually no observable change in the jet. Despite the liquid is in boiling conditions, the evaporating flow is not large enough to disturb the surface of the jet. Since the liquid entering the chamber is fresh, the short time the liquid stays in the field of view is not enough to observe bubbles nucleating. However, for a fixed driving pressure there is a critical value of the pressure ratio P_∞/P_v for which some morphological changes appear close to

the nozzle. First, the initially axisymmetric jets exemplified on Fig. 2(b-g) evolve to the jets with irregular cross section seen Fig. 3 and 4(a). Fig. 3 shows the temporal evolution of a jet of PFnH discharged through a metal nozzle of $d = 150 \mu\text{m}$. In this experiment we estimated a Jacob and a vapor Weber numbers of $Ja = 44$ and $We_v = 7.5$. These parameters are plotted as an orange cross on Fig. 1. The irregular cross section of the jet evolved due to the increased evaporation from the reduced ambient pressure. This can be observed as a change in the light refraction through the jet. This change in morphology usually marks the onset of flash-boiling. Close to the nozzle, the meandering jets do not seem to flash. However, we observed jets flashing far from the nozzle under the same conditions when widening the field of view. This entails that the penetration length, *i.e.* the distance from the nozzle to the flashing point, changes dramatically with relatively small fluctuations of the pressure around the critical value.

Figure 3: Temporal evolution of a jet of PFnH. The inner diameter of the nozzle is $d = 150 \mu\text{m}$, the pressure ratio is $P_\infty/P_v = 93 \times 10^{-3}$.

3.1.2. Onset of bubble nucleation

In Fig. 4 we show the onset of flash-boiling recorded from a video clip at one million fps. Here, a jet of PFnH is discharged from a metal nozzle with inner diameter of $100 \mu\text{m}$. In the experiment the vapor Weber number was fixed to $We_v = 3.5$ and the pressure ratio is $P_\infty/P_v = 0.18$, *i.e.* about 5 times smaller than the vapor pressure of PFnH. The pressure ratio in combination with the thermodynamic properties on Table. 2 result in an estimated $Ja = 33$. The dimensionless parameters

Figure 4: Single bubble explosion on a PFNH-jet. (a-f) The temporal evolution of a single bubble expansion at $P_{\infty}/P_v = 0.18$. The inner diameter of the nozzle is $d = 100 \mu\text{m}$, the total driving pressure is $P_d = 153 \text{ kPa}$. The images were recorded at 1 Mfps. The video of this experiment is included as additional material.

corresponding to this experiment are plotted as a yellow square in Fig. 1. While some sections break due to the nucleation of large bubbles, the rest of the jet continues moving upwards at the same speed it was initially discharged. At this moderate vacuum, we only observe the nucleation of large bubbles that usually break the jet in large sections. Smaller nuclei probably do not have enough energy to expand at this point, taking longer times than our recordings to achieve the critical size for a violent expansion. Some time after the onset of expansion, the liquid sheet separating the vapor of the bubble and the ambient gas becomes very thin. At this point the bubble bursts when a small hole nucleates on its surface. The retracting film forms a rim which breaks up into droplets that

are ejected in all directions. The Jacob number corresponding to this experimental conditions is smaller than the prediction given by Eq. 2 when bubbles start to nucleate, *i.e.* $Ja = 46$. Such difference may result from the uncertainty of exact physical properties of Flutec-PP1. Alternatively, it is also possible that jets break usually in these conditions. But, since a large portion of the jet remains undisrupted and moving upwards, this was not observed in previous reports.

Fig. 5 shows the MPIP calculated from Fig. 4, which reveals the trajectories of the drops produced after the bubble bursts. This figure reveals that drops formed during flash-boiling actually expand in wider spray angles than previously re-

Figure 5: The trajectories of the droplets ejected after the expansion and rupture of the bubble. This image was obtained with a MPIP from the video recording in Fig. 4.

ported. Similar and subsequent bubble expansions appear when the flash-boiling instability starts and they are responsible for the overall spray angle.

3.1.3. A larger surface enhances heat transfer

It is possible that, once the jet has been sectioned by the burst of a bubble, the heat transfer is locally augmented with a greater surface exposed. Thus, the recently created interphase becomes even more susceptible to bubble nucleation, as observed in Fig. 6. The liquid used in this experiment was PFnH and it was ejected from a nozzle used with inner diameter of 150 μm . The vapor Weber number of the experiments performed with these nozzle was estimated as $We_v = 7.4$, as seen on Table 4. The bubble expanding in Fig. 6 exhibits characteristic lines across its surface. The lines may result from a change in the refractive index of the liquid, occurring due to a local variation in the density. The changes in the density of the liquid may occur due to inhomogeneous evaporation. In Fig. 6(a), a new bubble starts expanding on the lower part of a broken jet. Due to the large area exposed, the bubble grows very rapidly to a size many times larger than the diameter of the jet. The experimental conditions in Fig. 6 ensured the interphase of the jet would remain unperturbed at normal ambient pressure. For the corresponding vapor Weber number, Eq. 2 predicts that bubbles start nucleating on the jet if $Ja > 41$. In Fig. 6 we estimated a Jacob number slightly larger than this critical value, *i.e.* $Ja = 45$. Therefore, this experiment corresponds to a

regime where the first bubbles appear.

3.1.4. Developed Flash-boiling

As ambient pressure is further reduced, the number of simultaneous bubbles nucleating at the interphase increases drastically. The temporal evolution of a MeOH jet with a large number of daughter bubbles is shown in Fig. 7. The images of Fig. 7 were recorded at 2 Mfps. A metal nozzle with an inner diameter of 200 μm was used to discharge the liquid at a pressure ratio of $P_\infty/P_v = 2 \times 10^{-3}$. The flow conditions correspond to a vapor Weber number of $We_v = 3.9$. Using the thermodynamical properties of Table 2, we estimated a Jacob number of $Ja = 295$. This value is larger than the critical Jacob number required for a fully flashing jet predicted by Eq. 2, $Ja > 123$. The dimensionless parameters have been plotted as a green circle in Fig. 1. Large bubbles are observed nucleating at the interphase far from the nozzle in Fig. 7. These bubbles expand further than the single bubble in Fig. 4. Multiple bubbles nucleate in some regions of Fig. 7(c). Multiple nucleation usually results in faster expansion velocities than those for single bubbles. This occurred due to bubbles nucleating on the strained liquid surface around pre-existing bubbles. A possible explanation of the multiple nucleation is the enhanced heat transfer on the larger surface area previously described. A large number of small bubbles are nucleating at close distance from the nozzle in Fig. 7. Unlike in PFnH jets, small bubbles do not break the MeOH jet in sections. Remarkably, the bubbles nucleate smoothly on the free surface, leaving the inner core of the MeOH jet almost unperturbed. Possibly, the higher surface tension of MeOH results in a more stable film retraction. Unlike the bubbles nucleating in PFnH observed in Figs. 4 and 6, MeOH bubbles burst differently than PFnH bubbles as well. After a hole nucleates on its surface the largest bubbles seem to deflate and the film retracts rather than breaking violently, as seen in the upper part of Fig. 7(f-i). Possibly, the higher surface tension of MeOH results in a more stable film retraction. Such deflation occurs mostly for larger bubbles. A bubble of greater size indicates that it had initially more liquid available to expand. Therefore, the liquid wall of larger bubbles is presumably thicker than for small ones. The type of expansions observed in MeOH contrast with what was observed in PFnH. In PFnH the

Figure 6: (a-f) Temporal evolution of a **PFnH** jet previously sectioned. The broken jet exposed a large area where a large bubble forms. This bubble grows larger than the diameter of the nozzle and exhibits a clear change in its refractive index. $P_{\infty}/P_v = 89 \times 10^{-3}$ and the total driving pressure was 130.1 kPa. The frame rate was 2Mfps.

burst of the bubble sections the jet completely, forming tiny droplets very efficiently.

3.1.5. Pinch off, bursting and the thickness of the liquid layer

Images of a small bubble nucleating, expanding and bursting on a MeOH jet are shown in Fig. 8. The video recorded at 2 Mfps, focuses closely on the side of a jet discharged at a pressure ratio of $P_{\infty}/P_v = 2 \times 10^{-3}$. The conditions in Fig. 8 are similar to the experiment in Fig. 7, as shown in the dimensionless parameters reported on Fig. 1. Large bubbles form and burst in MeOH in a different way than small bubbles. Small bubbles keep a more rounded shape during the expansion. After a small bubble bursts within MeOH, the breakup of

the thin sheet is as violent as the large bubble of PFnH, shown in Fig. 4. These similarities in the behavior of bubbles of different sizes in different liquids can be explained through the differences in the physical properties of the liquids. Besides, the film forming the small bubble in Fig. 8 is much thinner than the film of bubbles seen on the top part of Fig. 7. The surface tension forces drive the faster film retraction on the small bubble of Fig. 8. Here, the retraction occurs faster due to a thinner film. This interpretation also explains that the bubbles at the top of Fig. 7 seem to deflate, as the retraction occurs slowly.

During the lateral expansion of the bubble, the surface at the opposite side of the jet is strained more than the surface closer to the core of the jet.

Figure 7: Temporal evolution of a flash-boiling jet of MeOH. (a-i) Show an increasing amount of bubbles nucleating on the surface of the jet. (g-i) The bubbles burst, breaking into droplets. The jet is ejected from a metal nozzle with an inner diameter of 200 μm for $P_\infty/P_v = 2 \times 10^{-3}$. The total driving pressure is 401 kPa. The images were recorded at 2 Mfps. The video of this experiment is included as additional material.

This results in a film with a non-uniform thickness, where the film is thicker close to the jet when compared to the tip of the bubble. At later stages of the hole growth, the film that initially formed the bubble retracts and breaks into tiny drops. Many of these drops are smaller than the resolution of our optical setup, $d_d = 7 \mu\text{m}$, making them difficult to see after the breakup. However, the growth velocity of the hole may provide an estimation of the smallest droplets formed. In Fig. 8, we measured an initial velocity of the diameter of the hole opening at $42.5 \pm 3.6 \text{ m/s}$. The hole grows due to the elastic retraction of the liquid film. The Taylor-Culick model provides an estimation for the velocity of a liquid film retracting due to the surface tension and limited by inertia (Savva and Bush, 2009). Using the Taylor-Culick (Thoroddsen et al., 2006, 2011), this velocity can be used to estimate the wall thickness h of the bubble immediately before the hole breakup. The Taylor-Culick equation is

$$h = \frac{2\sigma}{v_{TC}^2 \rho l}, \quad (3)$$

where v_{TC} is half of the growing velocity of the diameter of the hole. On the experimental conditions of Fig. 8, this model estimates a film thickness of $h = 129 \pm 21 \text{ nm}$. This results is not unique, but consistent with other measurements of MeOH, where half of the retraction velocities ranged from

11 up to 23 m/s. This translates to an estimated film thickness varying from $h \in [108, 471] \text{ nm}$. Using the growth velocities of the hole and the corresponding film thicknesses, we can estimate the Ohnesorge number (Oh). The Ohnesorge number quantifies the relationship between the viscous with the inertia and surface tension. For a retracting liquid sheet, Savva and Bush (2009) defined the Ohnesorge number as, $\text{Oh} = \mu/\sqrt{2h\rho l\sigma}$. We estimated this Ohnesorge number in the range of $\text{Oh} \in [0.1, 0.3]$. This range of Oh correspond to an intermediate regime where a rim forms (Savva and Bush, 2009). This was experimentally confirmed on the images of Fig. 8. They reported that the size of the rim may vary between four and nine times the film thickness. The joint between the rim and the film concentrates the stress, having the greatest potential for rupture. This results in the formation of drops with diameters ranging from 432 nm to $4\mu\text{m}$, according to Eq. 3. These drops are too small to be seen with our optical resolution. However, the formation and breakup of the rim is observed in Fig. 8. These results indicate that some drops are being formed on the sub-micron range. It also suggests that if the liquid sheet is unstable, there might be a few drops being formed within the hundreds of nanometers range.

Figure 8: Temporal evolution of a single bubble expanding on the surface of a MeOH jet. After expanding to its maximum size, the bubble is pinched off and breaks into droplets. The jet is ejected from a metal nozzle with an inner diameter of 200 μm . $P_\infty/P_v = 2 \times 10^{-3}$. The total driving pressure is 401 kPa. The images were recorded at 2 Mfps.

3.2. Spray Angle

One of the most common measurements in any controlled atomization is the spray angle θ_s . In general, the spray angle is highly correlated with the stability of the jet, regardless of the origin of the breakup mechanism. To distinguish between the mechanical and the thermal effects we trigger these mechanisms independently. First, we recorded the evolution of the jet while systematically varying the driving pressure with the vacuum chamber open to the atmosphere ($P_\infty \approx 101.325$ kPa). The video recordings were processed using the MPIP technique revealing the spray angles shown in Fig. 9(a). We performed experiments discharging different liquids through nozzles with different inner diameters. The liquids used were PFnH, MeOH and EtOH, discharged through metal nozzles with d from 100 to 200 μm . The measurements of the spray angles are summarized in Fig. 9(b). The liquids and the inner diameters of the nozzles are displayed with different symbols. The spray angles increased from 0 to less than 35° with a variation in the Reynolds number from 0 to 7000. Around $\text{Re} = 3500$ there is a transition from angles smaller than $\theta_s < 5^\circ$ to values slightly larger than 20°. This transition is clearly marked as a function of the Reynolds number, contrary to the most obvious Weber number. To compare the evolution of the spray angle, some of the

measurements were performed 2 cm downstream the nozzle outlet. Far from the nozzle outlet the measurements resulted in similar spray angles.

3.3. Reducing P_∞/P_v for small We_v

After characterizing the influence of the Re on the spray angle at ambient pressure, we explored the influence of a low pressure environment by varying only the pressure ratio P_∞/P_v . To do this, we calibrated the velocity of the jet as a function of the driving pressure. In this way, we avoid disturbed surfaces and meandering jets. In the following set of experiments, we decreased the ambient pressure while keeping a small We_v . This allowed to investigate the transition from a cylindrical and a laminar jet to a fully flashing jet. In Fig. 10(a) we show a jet of PFnH being discharged through a metal nozzle of 100 μm of inner diameter. The images of Fig. 10 were recorded at 200 kfps and a pressure ratio of $P_\infty/P_v = 10^{-3}$. We observe that the atomization is strong enough to fully disrupt the jet and atomize it into droplets. These drops are being ejected in all directions, as can be seen in the MPIP of Fig. 10(b). This pattern of trajectories was observed at a vapor Weber number of $We_v = 3.6$. Using the thermodynamical properties of PFnH in Table 2, we estimated $\text{Ja} = 98$. According to Eq. 2, bubbles disrupt the jet when the $\text{Ja} > 46$,

Figure 9: (a) Spray angles for different Reynolds numbers estimated after the MPIP. The experimental conditions do not appear on Fig. 1. (b) Spray angle as a function of the Reynolds number for different nozzles and liquids.

while the fully flashing regime is reached if the $Ja > 125$. This Jacob number corresponds to a regime where bubbles affect the jet but fully flashing is still not developed. The spray angle was calculated with the minimum and the maximum angle of the trajectories as marked in Fig. 10(b). The spray angle calculated in this way reflects much better the possible directions of the drops than previous reports of the spray angle.

The distribution of the pathlines in the angular direction θ , can be obtained from Fig. 10(b), by taking the average of the MPIP in the theta direction. The line $\theta = 0$ is defined on the direction of the PFnH jet. In Fig. 11, we present measurements of pathline distributions for three different ambient pressures. The red triangles of Fig. 11 represent the average angular pixel intensity of the perfectly cylindrical jet on Fig. 2(f). This experiment was

performed at normal ambient pressure, where $P_\infty/P_v = 4$. A top-hat distribution with a very small standard deviation, shown as a fine broken line in Fig. 11, fits the best to describe the jet. Using this distribution we calculate that normalized pixel intensity is reduced to 0.5 when the angle achieves $\theta = 4^\circ$. When the pressure ratio is reduced to $P_\infty/P_v = 9.2 \times 10^{-2}$, we observed the single bubble explosions in Fig. 6. In this case, the yellow diamonds in Fig. 11 are the calculated average pixel intensity in the angular direction. The increased decompression reshapes the path lines from a symmetric to asymmetric distribution. The two gamma functions at both sides of the jet are shown as the coarsely broken line of Fig. 11. The asymmetry results from a single expansion on the left side of the jet, making the distribution wider on one of the sides. On Fig. 6 there are more drops moving on the side where the bubble nucleated. In this case, the normalized pixel intensity is reduced from 1 to 0.5 when the angle achieves $\theta = 20^\circ$ and $\theta = 39^\circ$ on the left and right side, respectively.

Contrary to the prediction of Eq. 2, complete flashing has developed at $P_\infty/P_v = 10^{-3}$. We observe in Fig. 12(a) that the angular distribution of the pathlines conforms to the Gaussian function. The blue circles in Fig. 11 are the average angular intensity calculated from Fig. 10(b). The continuous line is the fit of the pathlines to a normal distribution. The normalized pixel intensity is reduced from 1 to 0.5 when the angle achieves $\theta = 57^\circ$. When complete flashing develops, individual contributions from each bubble explosion are averaged and the angular distribution becomes axisymmetric, dispersing liquid in all directions. This entails that the phenomenon at lowest vacuum occurs in a much smaller time and length scale than when single bubbles nucleate. The size and the velocity of droplets ejected by the flashing jet provides also relevant information.

We used particle tracking to measure velocities of the individual droplets ejected in the experiment shown in Fig. 10(b). The distribution of the velocities of 1045 droplets is shown in Fig. 12(a). The jet velocity was calibrated as a function of the total driving pressure in separate experiments at ambient pressure. At ambient pressure the velocity of the jet in Fig. 10(b) is 5.6 m/s. Such velocity corresponds to the mode of the velocity seen in

Figure 10: (a) Example of a frame showing a jet of PFnH flash-boiling. The frame rate was 200 kfps, the $d = 100 \mu\text{m}$ and $P_\infty/P_v = 10^{-3}$. (b) MPIP of the same recording showing the distribution of the pathlines. The video of this experiment is included as additional material.

Figure 11: Distribution of PFnH jets vs θ for different values of the pressure ratio P_∞/P_v .

droplets ejected in the histogram of Fig. 12(a). The velocity of the jet is shown as a solid green line in Fig. 12(a, b). Thus, there is a large number of droplets moving at the same speed of the jet. However, the average of the velocity is 6.56 m/s. So, the overall velocity of the drops ejected is more than 17% larger than the jet velocity. Therefore, the atomization mechanism adds additional momen-

tum to the liquid. In fact, the gamma distribution of Fig. 12(b) is skewed towards larger velocities, reaching even 10 m/s when the probability is 0.05. Measuring the diameter of every drop tracked in the experiment of Fig. 10(b), we can correlate droplet size with its velocity. This correlation is shown in Fig. 12(b). The horizontal broken line marks the limit of the spatial resolution of about $7 \mu\text{m}$ in Fig. 10(b). The equivalent diameters of Fig. 12(b) were calculated from the area of the drops in the image, as $d_{eq} = 2(A/\pi)^{1/2}$. There are a large number of droplets that are smaller than this limit, as we mentioned when discussing Fig. 8. However, they are so small, we cannot determine their size or position accurately. In Fig. 12(b) the equivalent diameter shows a maximum when the velocity is around 5 m/s, i.e. close to the jet velocity.

3.4. Taylor-Culick model

We compare the experimental correlation of Fig. 12(b) with the Taylor-Culick velocity (Thoroddsen et al., 2006, 2011), which models the speed of retraction of a liquid film dominated by surface tension. This model was previously used in

Figure 12: (a) Probability distribution of the velocity of the drops ejected at $P_\infty/P_v = 10^{-3}$. (b) The equivalent drop diameter d_{eq} as a function of the velocity of the drop v_d .

Eq. 3 to estimate the thickness of the wall of the bubble. By solving Eq. 3 for the velocity we obtain

$$v_{TC} = \sqrt{\frac{2\sigma}{\rho h}}. \quad (4)$$

To test the validity of this hypothesis, we assumed that droplets form with a diameter similar to the thickness of the film. The result of this calculation is shown in the solid line of Fig. 12(b). According to this prediction, smaller droplets move faster than larger droplets. However, Taylor-Culick fails to predict the velocity of droplets larger than the spatial limit resolution, i.e. $7 \mu\text{m}$. Fig. 12(b) reveals that the largest droplets have velocities slightly smaller than the velocity of the jet (solid green line). Such velocity loss might occur due to energy dissipation during the jet breakup into small droplets. Even

when assuming the thickness of the film is one order of magnitude smaller than the equivalent diameter, the distribution in Fig. 12(b) does not compare with the prediction (broken line).

3.5. Spray Angles vs P_∞/P_v in PFnH

The intensity of the flash-boiling was characterized with the spray angle θ_s . Previously, we described the spray angle as a function of the Re in Fig. 9(b). But more fundamentally, the spray angle changes drastically as a function of the pressure ratio P_∞/P_v . We show measurements of the spray angle as a function of the pressure ratio for nozzles with different inner diameter in Fig. 13. The diameters were varied from 30 to $150 \mu\text{m}$ using glass and metal nozzles discharging PFnH. The range of inner diameters of the nozzles resulted in a variation of the We_v from 0.1 to 100. Despite the different roughness in the materials, we observed similar results for similar We_v . The wide spray angle measured in Fig. 10(b) is consistent with systematic measurements varying the pressure ratio while keeping a small jet velocity. Once the ambient pressure becomes smaller than the vapor pressure of PFnH ($P_v = 25 \text{ kPa}$), spray angles of similar magnitude are displayed. For most nozzles, the spray angles achieve values larger than $\theta_s > 300^\circ$, when the pressure has dropped below the vapor pressure of PFnH. The transition towards full vacuum occurs gradually for the metal nozzles. However, there are many realizations where the spray angle is zero. Similar results are observed for the glass nozzles, with the exception of the two smallest nozzles.

The spray angle of the two smallest nozzles (d of 30 and $45 \mu\text{m}$) does not rise above 100° , even for the lowest vacuum pressures in Fig. 13. With these two nozzles the vapor Weber number decreases to a value of $We_v = 0.1$, resulting in the inhibition of the flash-boiling mechanism. Such inhibition is a natural result of crossing the lower threshold given by Eq. 2, shown as the broken line of Fig. 1. Due to the weak dependance in this relationship, a large variation in the We_v results in a small variation in the critical Jacob number. In this case, a variation of 4 orders of magnitude in the We_v translated to a factor two change in the critical Jacob number (from 100 to 200) necessary to flash-boil. This increase in the threshold is enough to inhibit the flash-boiling in small nozzles. Consistently, we did not observe bubble expansion after the nozzle's

discharge in these experiments. However, using the smallest glass nozzle ($d = 30 \mu\text{m}$) the spray angles increased in vacuum pressures up to 30° . Here, the spray did not form from bubbles expanding and bursting. We therefore hypothesize that the sharp tip of the glass nozzle induces an instability on the jet only in vacuum conditions. The sharp geometry of the tip results from the method to produce the nozzles, *i.e.* glass pulling. In this case, the jet with a small We_v naturally needs higher thermal energy to enter the flash boiling regime, according to Eq. 2. At this stage the jet displays angles much smaller than in flash-boiling conditions, similar to Fig. 9(a). For the glass nozzle with $d = 45 \mu\text{m}$, the spray angles were close to 100° at the lowest vacuum. The expansion of bubbles was not observed in these experiments, although some parts of the jet burst. In this case, the jet broke immediately into drops when a bubble appeared on the surface of the jet. As a result, wider spray angles than the ones for the nozzle of $d = 30 \mu\text{m}$ were observed. Despite the greater spray angles, these were still much smaller than in a developed flashing jet.

Figure 13: Spray angle as a function of the pressure ratio P_∞/P_v . Glass and metal nozzles with inner diameters from 30 to $150 \mu\text{m}$ were used to discharge PFnH.

Nozzles with inner diameters larger than $45 \mu\text{m}$ displayed a clear trend of the spray angle vs the ambient pressure. In all the experiments using PFnH, the We_v was smaller than 100. In particular, the experiments with PFnH discharged through metal nozzles were performed at a fixed We_v number, *i.e.* 3.6 and 7.4 for an d of 100 and $150 \mu\text{m}$, respectively. Ambient pressures above the vapor

pressure of the liquid, resulted typically in spray angles close to $\theta_s = 0^\circ$. However, the spray angle continuously increased when the ambient pressure decreased below the vapor pressure of PFnH. The spray angle reached values $\theta_s > 300^\circ$ about one order of magnitude below the vapor pressure of PFnH ($P_v = 25 \text{ kPa}$), $P_\infty/P_v \sim 0.1$. As long as the total driving pressure was small, these results were similar using nozzles of different materials and inner diameters larger than $45 \mu\text{m}$.

Note that there is a large amount of realizations where the spray angle was measured close to zero. This occurs because the penetration length (the distance from the discharge to the flashing point) varies greatly very close to the critical pressure required for flash-boiling. This sometimes resulted in flash-boiling occurring beyond the field of view of the camera. Nevertheless, the large amount of experiments performed unraveled the trend of increasing θ_s as the ambient pressure is reduced. Notice the development of the spray angle is probabilistic rather than a deterministic measurement. Its value depends also on other factors, besides the variations of the penetration length. The individual trajectories of the drops have a major influence on the spray angle. The density of ejected drops is drastically reduced in the opposite direction of the jet. Since there are only a few drops moving in these direction, the spray angle may vary considerably if only a few drops were not ejected during the recording. In other words, due to the low density of the spray at wide angles, drops moving in this region have a mayor impact on the spray angle. Despite this large spread of the data, the maximum spray angle drastically increases when the ambient pressure is reduced below the vapor pressure of the liquid, as seen in Fig. 13. Some experimental points are not being displayed in Figs. 13 and 14, to avoid overlap of the points.

3.6. Spray Angles vs P_∞/P_v in other liquids

We also measured the spray angles of three other liquids, MeOH, EtOH and 1-PB. The results are summarized in Fig. 14 as a function of the pressure ratio P_∞/P_v for different nozzles. The filled symbols correspond to metal nozzles and the empty symbols correspond to glass nozzles. In Figs. 13 and 14, the spray angles achieve values $\theta_s > 300^\circ$ for pressure ratios P_∞/P_v smaller than

one. This pressure ratio is achieved when the pressure within the chamber is reduced below the vapor pressure of the liquid. In this section the nozzles have a larger inner diameter, necessary to discharge liquids with higher surface tension. As seen in Fig. 7, a liquid with a larger surface tension is more difficult to break into sections by the expansion and breakup of bubbles. Remarkably, the bubbles produced by these jets are larger than with PFnH. However, these morphological differences do not influence the spray angle as it only depends on the drop trajectories.

Figure 14: Spray angle as a function of the pressure ratio P_{∞}/P_v for MeOH, EtOH and 1-PB. The open and filled symbols refer to glass and metal nozzles, respectively.

Despite the vapor Weber number in Figs. 13 and 14 is on a similar range, $We_v \in [0.1, 10]$, the change in θ_s seems to be more abrupt. A possible explanation for the sharper increase might lay on the fact that larger inner diameters result in a smaller range of velocities where the surface of the jet remains unperturbed. This leads to a narrower range of driving pressures that produce a jet with an undisturbed surface. The larger inner diameters were necessary to allow using liquids with higher surface tensions than PFnH.

The least volatile liquid is EtOH, with the lowest vapor pressure in Table 3. Its low volatility in combination with its slightly higher viscosity results in jets with a very stable surface, that are therefore very difficult to flash-boil. This is consistent with the spray angles shown in Fig. 9(b), where the maximum Reynolds number of EtOH is below 1000. A jet with a stable surface combined

with a liquid with a low volatility results in no flashing for the two smallest nozzles ($d = 66$ and $100 \mu\text{m}$). In all the other cases, MeOH, EtOH and 1-PB achieve large spray angles when the ambient pressure is reduced below the vapor pressure of the liquid. The values of the vapor pressures of these liquids can be found on Table 3.

3.7. Bubble expansion

Besides measuring the spray angles, the ultra-high speed camera allowed observing the expansion of individual bubbles. In flash-boiling conditions, bubbles usually start forming near the free surface of the jet, as seen in Figs. 4, 6, 7 and 8. The core of the jet is an obstacle for the expansion and therefore bubbles usually expand the most when they are formed close to the outer edge of the jet. For purely lateral bubble expansions, we measured the distance from the tip of the bubble to the undisturbed jet surface as a function of time, x_e . An example of this distance is exemplified with a blue arrow in Fig. 8. The derivative of the distance x_e as a function of time is the expansion rate of the bubble, v_e . In most cases, the derivative was estimated as the derivative of a polynomial of 4th degree fitting x_e . Examples of these measurements as well as the polynomial fittings are presented in Figs. 15(a-d). In these experiments PFnH is discharged through metal nozzles of $d = 100 \mu\text{m}$. The pressure ratio is $P_{\infty}/P_v = 10^{-3}$. The most common evolution of the expanding distance is presented in Fig. 15(a). At the beginning the bubble grows slowly in size. As time passes, its growth rate is dramatically increased. The low section of Fig. 15(a) shows the corresponding increase on the expansion rate of the bubble. The measurement of the expansion distance x_e is stopped immediately before the bubble bursts. The growth rate in this example starts at a zero velocity and achieves up to 60 m/s in less than 25 μs . Such a dramatic change in the velocity requires an exceptional acceleration of $10^5 g$.

Another typical bubble expansion is shown in Fig. 15(b). The bubble seems to expand at a constant rate at the beginning. Then the driving forces stop and the growing bubble decelerates, ultimately reaching a steady size. As we see in the broken line of Fig. 15(b), the velocity decreases continuously from the beginning of the expansion. More complex scenarios of bubble expansion occur

in Fig. 15(c) and 15(d). A bubble expands slowly at the beginning of Fig. 15(c). Then, the velocity increases exponentially, similar to the scenario of Fig. 15(a). However, the velocity decreased again, ultimately reaching zero velocity.

Besides these, some complex scenarios arise from the nucleation of many bubbles in the same region. In these cases, the expansion velocity is the result of many adjacent expansions that might start one after another. These very complex scenarios result in unique bubble expansions that cannot be understood as a single event but from the addition of sequential or simultaneous events. Such is the case of Fig. 15(d), where the expansion displays cycles of acceleration and deceleration, within a very short time span.

Compiling numerous experiments, we calculated the maximum expansion velocity v_e^{max} as a function of the pressure ratio P_∞/P_v . The measurements for PFnH are shown in Fig. 16, while Fig. 17 shows results for other liquids. The inner diameter of the nozzles in Fig. 17 varies from 140 to 480 μm . The maximum velocity v_e^{max} was calculated as the maximum value for v_e on individual bubbles during the interval recorded. For ambient pressures lower than the vapor pressure of the liquid, the maximum velocity of expansions v_e^{max} increases continuously as P_∞ is lowered. The increase in the maximum velocity is similar to the one observed for the spray angle of Fig. 13. At the smallest pressure ratio of Fig. 16, the maximum expansion velocities reach close to 140 m/s . These experiments were performed with metal and glass nozzles ranging from 68 up to 150 μm . When the pressure is further reduced below the vapor pressure of the liquids, the expansion rate of the bubbles increases. The maximum expansion rates reported for PFnH in Fig. 16 are of the same order of magnitude as the velocities in Fig. 17, where at the highest vacuum the expansion rate achieved is about 120 m/s .

4. Discussion

4.1. Spray angles

Previous studies have defined the spray angle as the distance the jet has opened at some fixed length from the nozzle. For non-evaporating sprays this angle reflects the directions the liquid takes during the atomization. However, for a

flash-boiling jet the strong evaporation results in jets that look parabolic in the images inasmuch as the angle is not constant. Besides, most of the previous studies have made recordings at lower frame rates than 13,000 fps. This results in pictures with motion blur that can only be translated to average pixel intensity through an intensity threshold. These factors result in an intrinsic under estimation of the direction the liquid and the gas take. Furthermore, the average intensity profiles depend on the exposure time and the illumination. Therefore, measurements are only comparable if the illumination conditions are the same. This has a great impact when trying to compare measurements from different experimental studies, as the results are usually not comparable. This is the main reason why the spray angles reported in the literature are very different between one another.

Notice that in our experiments the spray angle is calculated using approximately 166 frames. If these frames were recorded at 5 million frames per second, the total time of the recording is 33 μs . Such a short recording time also presents a limitation regarding the information that can be acquired. For example, this measurement of the spray angle depends on the maximum and the minimum angle of the ejected droplets. The presence of these droplets is intrinsically probabilistic and the chances of larger angles increase with the total recording time. However, we believe that spray angles accounted in this way reflect much better the direction the droplets are taking. The MPIP of Fig. 7 was calculated from 166 frames recorded at 2 Mfps . This results in a spray angle calculated over 83 μs . For a typical high-speed camera recording at 12 kfps , this time scale would have elapsed in a single frame. Then, the spray angles measured with the ultra-high speed video clip are comparable with the instantaneous angles on the frames of a typical high speed camera.

4.2. Droplet velocity and size

We have demonstrated that flash-boiling produces droplets in a range of sizes, as observed in Fig. 12(b). This contravenes the common notion that flash-boiling results in drops with a narrow size distribution. In our experiments, the largest droplets have similar sizes to the inner diameter of the nozzle, i.e. 100 μm in Fig. 12(b). Since the

Figure 15: Typical bubble expansion in a PFnH jet under flashing conditions. $\circ$ marks the horizontal position of the bubble's boundary. The solid line represents the polynomial fitting and n its degree. The broken line represents the velocity calculated from this fit. (a) Most common bubble expansion. (b) Bubble with a fast initial expansion, reducing speed as times progresses. (c) A bubble with an expansion speed that initially increases, reaches a maximum and then decreases. (d) A complex case of bubble expansion.

spatial resolution is limited to $7\mu\text{m}/\text{pix}$, we did not observe smaller droplets than that. However, some droplets **could** have sizes of the same order of magnitude as the film thickness of the expanding bubbles (Savva and Bush, 2009). For the MeOH jet in Fig. 8, the thickness of the bubble wall was estimated down to 108 nm from the Taylor-Culick model. However, it is very likely that most of the drops formed due to the breakup of the liquid rim after the pinch off of the bubble.

Besides, the range of droplet velocities also extends in different scales. As we observed in Fig. 12(b) there are droplets with a velocity smaller than 1 m/s . While, the measurements of the velocity of retraction could result in droplets moving up to 45 m/s . This is a variation of almost two orders of

magnitude in the droplet velocity.

4.3. Considerations on the prediction of the film thickness

According to the Taylor-Culick model, the thickness of the film of bubbles within MeOH jets has been estimated between 108 and 477 nm . For such a small thickness it is not obvious whether the assumptions that led to the Taylor-Culick model are still valid. Here we clarify some of these assumptions. The retraction of a liquid sheet has been studied in three different regimes of Ohnesorge numbers (Oh), small, medium and large (Savva and Bush, 2009). The Ohnesorge is considered small if $Oh < 0.1$ and large if $Oh \gg 10$. The medium regime is defined in between

Figure 16: Maximum expansion velocities of bubbles on jets of PFnH as a function of the pressure ratio P_∞/P_v . Three glass nozzles as well as two metal nozzles of different diameters were used to discharge the liquid.

Figure 17: Maximum expansion velocities of bubbles on jets of different liquids as a function of the pressure ratio P_∞/P_v . Three glass nozzles as well as four metal nozzles of different diameters were used to discharge the liquid.

these extreme values. On small Oh numbers, a rim appears at the front of the retracting sheet. Capillary waves form in the vicinity of the rim, disturbing the surface of the liquid sheet. For the three regimes the Taylor-Culick model estimated the definitive retraction velocity of the liquid sheet (Savva and Bush, 2009). The definitive velocity was achieved for asymptotically long times. On our measurements of the retraction sheet velocity on MeOH jets, the Ohnesorge was comprehended between $Oh \in [0.3, 0.6]$. In these intermediate values, there are two possible estimations of the asymptotic long time. In the absence of viscous dissipation, the sheet would achieve the definitive velocity for times longer than the inviscid time scale, $\tau_{inv} = \sqrt{\rho_l h^3 / 2\sigma}$. If there is viscous dissipation, longer times are defined by the viscous time scale, $\tau_{vis} = \mu h / 2\sigma$. In our measurements of the growth velocity of the hole, the inviscid time scale was comprehended between $\tau_{inv} \in [5, 43]$ ns. Meanwhile the viscous time scale was comprehended between $\tau_{vis} \in [1.4, 6]$ ns. Despite not being exactly the same, both time scales are of the same order of magnitude. The typical time scales seen in Fig. 8 correspond to 3 μ s, or more than two orders of magnitude. Then, the evolution of the retraction sheet is long compared with both the inviscid and the viscous time scales.

The stability of such thin films is compromised by other factors besides the regime of the retracting liquid sheet. Liquid films formed in flash-boiling

conditions are therefore far from being stable. There are a number of mechanisms that could be responsible for their breakup. One of them is the capillary pressure acting due to the non-uniform thickness of the film. We can construct a characteristic time scale for the capillary action to break the liquid sheet using the capillary velocity and the thickness of the film. This time scale is $\tau_{Ca} = \mu h / \sigma$. For the film thicknesses we reported, the capillary time scale is extremely short being comprehended between $\tau_{Ca} \in [3, 13]$ ns. The bubble seen on Fig. 8 exists for approximately 30 μ s or about four orders of magnitude longer than the capillary time scale. Then, we must conclude that capillarity stabilizes the continuous sheet, while it is known to destabilize the edge of the free film (Deegan et al., 2007; Zhang et al., 2010; Roisman, 2010).

Another mechanism may affect the stability of films with very small thicknesses, the disjoining pressure. According to Gumerman and Homsy (1975), there is a critical aspect ratio of the thickness and the radius of supported circular films, γ_c , for which the disjoining pressure destabilizes the sheet. This aspect ratio can be estimated with two dimensionless numbers, $S = A / 6\pi a^2 \sigma$ and $P_\infty a / \sigma$, where S is the Scheludko number, A is the Hamaker constant and a is the radius of the circular film. Assuming this criterium can be extended to a liquid film with the geometry of

the bubble in Fig. 8, we then calculate the critical aspect ratio γ_c corresponding to the experiment of Fig. 8. The Hamaker constant for MeOH is $A = 40.32 \times 10^{-20}$ J (Birdi, 2002) and the maximum equivalent radius of the bubble is roughly $a = 500$ μm . The ambient pressure is $P_\infty = 2225$ Pa in the experiment of Fig. 8. Then, the dimensionless numbers become $S \approx 4 \times 10^{-12}$ and $P_\infty a / \sigma \approx 50$. These two dimensionless parameters correspond to a critical ratio on the order of $\gamma_c \sim 10^3$ (Gumerman and Homsy, 1975). Meanwhile the ratio of the film is comprehended between $\gamma \in [10^{-4}, 10^{-3}]$. Even the largest aspect ratio is five orders of magnitude smaller than the critical value. This is consistent with soap films where the aspect ratios are much larger. Therefore, films with these typical thicknesses and radii are not pinched by the disjoining pressure.

4.4. Taylor-Culick

A section of the Taylor-Culick prediction curve of Fig. 12(b) falls in a region below the spatial resolution limit. However, the section of the curve that falls in the visible region has a different trend from the prediction. Therefore, we believe that the relation between d_{eq} and v_d is given by a different mechanism than the liquid retraction. One plausible mechanism is the difference between the internal pressure of the bubble and the ambient pressure. The inner pressure of the bubble is a combination of the vapor pressure of the liquid and gas pressure, whose ratio is unknown as it is beyond our measurement capabilities. Some clues might be found in the Rayleigh-Plesset equation, a proven model for cavitating bubbles. Brennen (2013) explained that the pressure inside a bubble is the addition of partial pressures of gas and vapor. One of them being the vapor pressure of the liquid and another being the pressure of the condensed gases. A liquid with such a low surface tension as PFnH is usually saturated with a large amount of gases. Since the total pressure inside the bubbles is unpredictable, a model for the bubble growth still eludes us.

4.5. Inertial bubble growth model

Fig. 16 shows that bubbles can expand within PFnH at speeds up to 140 m/s. Not only these are consistent with velocities reported in Fig. 17

for other liquids, but are exceptionally large when compared with typical examples of growing bubbles. This is why we consider appropriate comparing them with predictions for other rapidly growing bubbles. In this direction, Rayleigh-Plesset is a well accepted model for the growth and collapse of cavitating bubbles (Plesset and Prosperetti, 1977). This model estimates that the maximum expanding velocity of a bubble growing within an infinite pool of liquid (Prosperetti and Plesset, 1978) is

$$\dot{R} = \left[\frac{2}{3} \frac{P_v(T_\infty) - P_\infty}{\rho_l} \right]^{1/2} \quad (5)$$

Eq. 5 represents the upper bound velocity limit for a growing bubble limited by the inertia of the liquid. Even on a perfect vacuum ($P_\infty = 0$), the expansion velocity of a PFnH bubble estimated by this model is around $\dot{R}^{max} = 3$ m/s. This value is almost 47 times smaller than the expansion velocities we measured in Fig. 16. Such discrepancy results from the boundary conditions of the Rayleigh-Plesset equation. Namely, Eq. 5 was derived assuming that the bubble is nucleating in a infinite pool of liquid, which compresses the bubble more as it expands. The expansion velocity of a bubble predicted by Rayleigh-Plesset does not continuously increase. On the contrary, the diameter of the bubble will achieve a maximum value as the inner pressure of the bubble declines during the expansion, reaching zero velocity and ultimately collapsing. Other models for predicting the bubble growth rate relying on different driving mechanisms result in a further underestimation of the expansion rate. For example, a vapor bubble growing in a superheated liquid due to a heat flux (Prosperetti and Plesset, 1978) results in values much smaller values than 3 m/s. These mechanisms of bubble expansion are completely different from our experiments, where as the film gets increasingly thinner, the resistance to expand becomes weaker.

4.6. Divergent bubble growth model

To better estimate the bubble expansion velocity, we propose a model for a bubble expanding within a continuously thinning film. Our model only considers the inertia of the liquid opposing the expansion of the bubble. Such assumption naturally results in a divergent velocity. Let us assume there is a semi

Figure 18: (a) A semi spherical bubble nucleating on the side of a liquid jet. (b) The differential element of volume and area in of the semi spherical bubble.

spherical bubble growing on the side of a liquid jet, as shown in Fig. 18(a).

The outer and inner radius of the bubble are $r(t)$ and $r_i(t)$, respectively. The volume of the liquid film of the semi spherical bubble is

$$V = \frac{2\pi}{3} [r^3 - r_i^3]. \quad (6)$$

The thickness of the liquid film on the surface of the growing bubble is $h = r - r_i$. Notice that h , r_i and r are functions of time. Substituting $r_i = r - h$ in Eq. 6, we obtain

$$V = \frac{2\pi}{3} [r^3 - (r - h)^3]. \quad (7)$$

If the evaporation rate is small compared with the expansion rate, we can assume the volume V on Eq. 7 is constant. This assumption is supported by dimensional considerations as is further discussed at the end of the section. Taking the time derivative of Eq. 7 we obtain the following relationship between the thinning velocity $\dot{h}$ and the expansion velocity $\dot{r}$

$$\dot{h} = h \frac{2r - h}{(r - h)^2} \dot{r}. \quad (8)$$

Assuming that $r \gg h$, simply transforms Eq. 8 to

$$\dot{h} = \frac{2h}{r} \dot{r}. \quad (9)$$

Now, let us focus on the differential volume element of Fig. 18(b). The second Newton law can be written as

$$(P_i - P_\infty) dA = \frac{d}{dt} (\rho \dot{r} dV), \quad (10)$$

where P_i is the inner pressure of the bubble, dA and dV are the differential elements of area and volume. From Fig. 18 we see that $dA = r^2 \sin \theta d\theta d\phi$ and $dV = hr^2 \sin \theta d\theta d\phi$. P_i is the inner pressure inside the bubble and P_∞ is the ambient pressure. Substituting the area and volume elements in Eq. 10, assuming a constant density and cancelling the angles we obtain

$$\frac{P_i - P_\infty}{\rho} r^2 = \frac{d}{dt} (r^2 h \dot{r}), \quad (11)$$

which can be expanded by deriving with respect to time to the following relationship

$$\frac{P_i - P_\infty}{\rho} = \frac{2h}{r} (\dot{r})^2 + \dot{h} \dot{r} + \ddot{r} h. \quad (12)$$

Eq. 12 can be simplified using the result from Eq. 9 to

$$\frac{P_i - P_\infty}{h\rho} = 4 \frac{(\dot{r})^2}{r} + \ddot{r}. \quad (13)$$

For a gas bubble expanding within a constant volume of liquid, its radius r increases while its thickness h decreases. Therefore the variables h and r are entangled. To solve Eq. 13, we propose an additional relation between h and r as follows. Since the time scale of the bubble growth is much smaller than the evaporation, it is fair to assume that the initial volume of liquid around the bubble is conserved. In an ideal jet where there is no relative motion around a nucleation site, there is a limited amount of liquid in its surroundings that can be used for the bubble to expand. This volume can be quantified as

$$V_L = \frac{4\pi}{3} (r_o^i)^3, \quad (14)$$

where V_L is the volume of liquid available for the bubble to grow and r_o^i is the radius of that sphere. Assuming the vapor bubble has formed, the volume of the expanding sphere can be quantified from Fig. 18(a) as

$$V_L = Ah = 4\pi r^2 h. \quad (15)$$

Since the volume is conserved, both sides of Eqs. 14 and 15 can be equated, resulting in

$$h = \frac{(r_o^i)^3}{3r^2}. \quad (16)$$

Eqs. 16 and 13 can be equated and solved for the acceleration, resulting in a non-linear ordinary differential equation of second order,

$$\ddot{r} = \frac{3\Delta P}{\rho (r_o^i)^3} r^2 - 4 \frac{(\dot{r})^2}{r}. \quad (17)$$

A numeric solution of Eq. 17 using r_o^i and the initial radius $r(t = 0)$ as adjustable parameters is presented as a solid blue line in Fig. 15(a). The inner pressure P_i and the density ρ of the equation are the vapor pressure of the liquid and the liquid density of PFnH. The blue curve in Fig. 15 was calculated using the values of $r_o^i = 10.15 \mu\text{m}$ and $r(t = 0) = 0.45 \mu\text{m}$. The model preserves the characteristic divergent velocity of the expanding bubble. However, the concavity of the blue line is clearly larger than the experimental measurements.

Taking the values resulting from Eq. 17 that best fitted the experiment of Fig. 15(a) we can verify whether the assumptions underlying Eq. 7 are correct. The characteristic time scale of evaporation can be estimated from a balance between the heat needed to evaporate the liquid and the heat flux going through the wall of the bubble via conduction. A spherical portion of liquid with radius r_o^i is initially surrounding a nucleation site. The volume of the sphere V_L is calculated with Eq. 14 and its mass is calculated as $m = \rho_L V_L$. For a spherical volume of radius r_o^i surrounding the nucleation spot, the mass calculation results in $m = 4\pi\rho_L(r_o^i)^3/3 = 7.45 \times 10^{-12} \text{ kg}$. The total latent heat needed to evaporate this amount of liquid is $Q_v = h_{fg}m/M$, where M is the molar mass of PFnH, 338 g/mol. The value of h_{fg} for PFnH is 31.7 kJ/mol, as previously stated. Therefore, the total heat needed to evaporate the liquid is $Q_v = 7.0 \times 10^{-7} \text{ kJ}$. Prosperetti and Plesset (1978) gave an estimation of the heat flux going from the liquid through the wall of the vapor bubble. In our case, the heat flux is going from the ambient gas to the liquid wall. By adapting their result we write $\dot{Q}_w = 4\pi(r_o^i)^2 k_{air} \Delta T / D_{air} t^2$, where t is the time, k_{air} and D_{air} are the thermal conductivity and diffusivity of the air. These properties take the values of $k_{air} = 26 \times 10^{-3} \text{ W/m}\cdot\text{K}$ and $D_{air} = 19 \times 10^{-6} \text{ m}^2/\text{s}$. Equating Q_v and $\dot{Q}_w$ and solving for the time t , we obtain $t = r_o^i \sqrt{4\pi k_{air} \Delta T / D_{air} Q_v} \approx 16 \text{ s}$. Thus, the time for the spherical region surrounding the nuclei to evaporate is more than five orders of magnitude longer than the typical expansions measured in our study.

Furthermore, convection rather than conduction might result in a greater heat flux towards the surface. The velocity of propagation of evaporative fronts has been discussed before (Avksentyuk,

1995; Pavlenko and Lel, 1999; Pavlenko and Surtaev, 2010; Pavlenko et al., 2011; Zhukov et al., 2017). As a result of these investigations, it was concluded that the velocity of the evaporative front is dominated by the convective heat flux. However, these models have been developed for flat liquid films and strongly rely on the thickness of the thermal boundary layer. In these investigations the thickness of the boundary layer may be estimated from assumptions from the ambient conditions. Within the bubbles of our experiments, the evaporative flow remains confined. Here, conditions such as temperature and pressure are uncertain, leading to unknown thickness of the boundary layer, not to mention the difference in geometry. Therefore, we are unable to give a prediction of the convective heat flux leading to the evaporative flow. For reference, in the most explosive visualization of flash-boiling, corresponding to Figs. 7 and 8, the Ja has a value of 295, corresponding to a temperature difference of $\Delta T = 240^\circ\text{K}$. Such a large increase in temperature corresponds to much greater values than in previous investigations.

As seen in Fig. 15(a), there is a fundamental difference between the model and the experiments. In the experiments the velocity of the bubble increases very rapidly, nevertheless it does not diverge as quickly as the velocity predicted by the model. The contrasting behavior is explained as follows. The model of Eq. 17 considers that only the inertia of the liquid opposes the expansion and as the film gets thinner this only decrease, resulting on a diverging velocity. In reality, the evaporative flow rate is unable to keep the inner pressure of the expanding bubble constant. With the inner pressure declining, the force driving the expansion of the bubble is reduced.

Please note that the model presented in this section does not pretend to estimate exactly the expansion rates. But, introducing the foundations underlying the mechanism of bubble expansion. Furthermore, there is a number of factors that remain unaccounted, besides the lowering inner pressure. In general the expansion rates are greatly affected by other multiple variables, *i.e.* gradients on the evaporation rate, a complex geometry or even the breakup of the inner wall. Another variable unaccounted on the model is the convective movement of the flow. Velocity gradients might influence the dynamics of expansion or the

thickness of the film.

4.7. PFnH vs LPG

Our main motivation to investigate this topic was to further understanding the risk of explosion involved in the leaks of LPG and LNG. However, there is an intrinsic risk in doing experiments with flammable liquids. LPG is a combination of propane and butane (Totten et al., 2003). When liquid, propane has a surface tension between 7 and 11 mN/m (Lin and Duan, 2003). Some physical parameters of LPG, such as the liquid density (496 kg/m³, according to Lin and Duan (2003)) and the vapor pressure of the liquid (1 MPa, according to Lin and Duan (2003)) are different from PFnH. A smaller density implies less resistance of the liquid to be accelerated at larger velocities. A liquid with a higher vapor pressure carries a stronger potential for the formation of a finer flash-boiling spray. Also, a similar low surface tension corresponds to a high probability of evaporation as well as a large amount of dissolved gas. For these reasons, the angular distribution of the pathlines in Fig. 10(b) is expected to be narrower, with LPG being dispersed in wider angles. Despite the higher vapor pressure of liquid LPG, the pressure ratios P_{∞}/P_v are on the same range as our experiments. However, the high pressure required to store liquid LPG also results in much larger driving pressures. A more uniform angular distribution of the pathlines when predicting the formation of explosive and flammable clouds.

4.8. Supersaturation

Common notion suggests that flash-boiling appears due to the violent evaporation occurring in the low pressure environment of our experiments. However, an alternative mechanism can result in the nucleation and expansion of bubbles after a decompression. This is called supersaturation and it occurs in liquids that contain a large amount of dissolved gases (Epstein and Plesset, 1950; Enríquez et al., 2014; Sheeran and Dayton, 2014). Perfluorocarbons such as PFnH contain a large amount of dissolved atmospheric gases. Dias et al. (2004) have reported that the expansion of bubbles has been altered by the amount of gases within the perfluorocarbon. For example, dissolved gases have demonstrated to increase the maximum diameter

of cavitation bubbles nearly by a factor of two (Sheeran and Dayton, 2014). In a decompression, the growth of bubbles is driven by the degree of supersaturation (Epstein and Plesset, 1950; Enríquez et al., 2014). Remarkably, LPG is a combination of propane and butane, whose ratio changes depending on the season (Totten et al., 2003). Under some circumstances the propane could be saturating the liquid butane, calling for supersaturation effects as well. Although this topic is out of the scope of this paper, this mechanism might play a major role in the bubble nucleation mechanism.

4.9. Driving pressures

We believe that evaluating the change in the spray angle under a low pressure environment for larger driving pressures would be very interesting. In particular, finding the width of the normal distribution in Fig. 11 as a function of the Reynolds or the Weber number is of great interest for the risk assessment. However, we observed there is no significant difference, even under the largest driving pressures we could reach in our facility, as seen in Fig. 9(b).

5. Conclusions

We visualized flash-boiling of jets of different liquids using an ultra-high speed camera. By systematically reducing the ambient pressure within the vacuum chamber, we observe the different stages that lead to a fully flashing jet. The material of the nozzles was irrelevant in the experiments. On initially cylindrical jets, we noticed the onset of boiling by observing at the change of the morphology of the jet as well as the development of meandering. For pressures lower than the vapor pressure of the liquid, single bubbles start nucleating and expanding at the surface of the jet. When the bubbles burst on PFnH the jets are broken into sections, while the core of the jet remains intact on MeOH. In both cases, the bubble burst resulted in the atomization of droplets. Remarkably, the large temporal resolution revealed that droplets are ejected in all directions around the jet. With larger areas exposed to an increased heat transfer, more bubbles nucleated. This bubbles could be larger and faster, as observed in

Fig. 4. At very low pressures bubbles of many sizes nucleated, forming droplets of several sizes as well. Using the MPIP we measured the spray angle as a function of the Reynolds number and the pressure ratio. This image analysis technique allowed us to quantify the distribution of the path lines in the angular direction. Using particle tracking, we measured the distribution of the drop velocities. We also quantified the equivalent diameter as a function of the drop velocity. The spray angles were measured based on the trajectories of the droplets, illustrating that maximum spray angles are much larger than what was estimated with averaged pixel intensity images. We show that there are typical expansion rates that result in high velocities and accelerations. In particular, the velocity of the most common type of bubble expansion strongly diverges as it approaches to bursting. However, multiple events of bubble expansions might occur simultaneously, giving a unique character to each expansion. The expansion velocity of the bubbles achieved an unprecedented velocity up to 140 m/s. Measurements were performed on different liquids, over a range of nozzles with inner diameters from 30 up to 180 μm . Using the Taylor-Culick equation, we estimated that the smallest droplets are formed due to the breakup of the rim during the burst of a bubble. The equation in combination with measurements on MeOH, predicted that the smallest droplets ejected are on the submicron range. Then the size of the droplets is widely spread, contrarily to the common notion of flash-boiling sprays.

From the point of view of fuel leaks, we estimated that the lower density of LPG and the higher driving pressure due to pressurized containers, would lead to a more turbulent interface that might expand more uniformly in the angular direction. We discussed that very well known models fail to predict the large velocities of expansion. We mentioned that the effect of saturated gases remain unaccounted for but it is nevertheless important.

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