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► To cite this version:

Omar Moussa, Dominique Tarlet, Patrizio Massoli, Jérôme Bellettre. Investigation on the conditions leading to the micro-explosion of emulsified fuel droplet using two colors LIF method. Experimental Thermal and Fluid Science, 2020, 116, pp.110106. 10.1016/j.expthermflusci.2020.110106. hal-02565105

HAL Id: hal-02565105 https://hal.science/hal-02565105

Submitted on 26 Feb 2021

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Investigation on the conditions leading to the micro-explosion of emulsified fuel droplet using two colors LIF method

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ABSTRACT

Biofuels in the form of water-in-oil (W/O) emulsions are a promising substitution for hydrocarbon fuels, mainly thanks to the micro-explosion phenomenon that occurs under strong heating. Micro-explosion is the atomization of a W/O emulsion drop into daughter droplets because of the water embedded droplets phase change. This atomization leads to the reduction of pollutants and unburnt hydrocarbons emissions. However, micro-explosion is not a systematic phenomenon: indeed, for some cases the water phase change is weak and leads to a partial atomization, called "Puffing". This paper relates a novel approach to better understand the occurrence of micro-explosion. It consists on the identification of the drop triggering the atomization, called the trigger drop, and measure its properties (temperature, size and position within the emulsion drop) thanks to non-intrusive optical diagnostics. Two colors laser induced fluorescence (LIF2C) is the main method used in this work for temperatures. These temperature measurements showed promising results of both quantitative and qualitative aspects. After the calibration steps and the method validation, it is applied for the temperature measurements of water droplets embedded in oil with the maximum temperature recorded around 130°C. Using these measurements methods, a correlation is found between quality of atomization (either micro-explosion or puffing) and the properties of the trigger drop (temperature, size and position). When one of these parameters is not optimal, it can indeed be compensated by the others.

Keywords: Fluorescence, biofuels, atomization, emulsions

Highlights:

- The atomization trigger of emulsified fuel drops is investigated
- Temperature measurements using LIF2C are carried out
- The trigger drop's temperature, size and position affect the atomization occurrence

Introduction

Most pollutants and harmful particles produced by humankind (NO_x, PM, CO, soot...) are generated by fuels combustion, in the area of transportation, heating systems, energy production.... Knowing the effects induced by these emissions on the atmosphere composition, the energy overconsumption led to tremendous damages to our health and planet [1-2].

Liquid fuels (either fossil or biofuels) are injected into the combustion chamber in the form of a spray of droplets. This is called the primary atomization. Previous works showed that smaller are the droplets the more complete is the combustion reaction and this is mainly due to a better air/fuel ratio [3]. However, by changing the physicochemical properties of the fuels, a secondary atomization can occur, generating even smaller fuel drops. This phenomenon is called micro-explosion and is specific to emulsified fuels, like water-in-oil emulsions [4-7]. Micro-explosion strongly relies on the difference in the boiling points of oil ([200-300] °C) and water (100°C). Under a strong heating, the water dispersed droplets undergo a phase change that breaks up the emulsion drop into child droplets (Fig.1). The emulsion drop atomization permits a cleaner and complete combustion thanks to the emulsion drop break up [4-8].



Figure 1: Schematic representation of a micro-explosion

One may observe that micro-explosion is characterized by its apparent random aspect. Sometimes the water embedded droplets phase change is not powerful enough to break the oil layer, leading to a partial break up called "puffing" [9-10]. Thus, the atomization can be defined by a statistical parameter called "occurrence of micro-explosion", defined as the ratio of number of optimal atomization and number of emulsion drops tested [11].

The present paper relates the experimental analysis of the micro-explosion on-set of water/ntetradecane emulsion drops in Leidenfrost regime. The aim of this work is the identification of the physical conditions leading to a successful micro-explosion. Thus, the drop whose phase change generates the atomization (trigger drop) is sought and the study of the influence of its main parameters (size, temperature and position within the emulsion drop) on the occurrence of micro-explosion is performed. The measurements were conducted using non-intrusive Laser Induced Fluorescence (LIF) for size and position measurements and 2-Colors LIF for the temperature measurements.

1. Theoretical background

1.1 Micro-explosion occurrence

Micro-explosion is defined as the fragmentation, in a very short amount of time [12] of an emulsified fuel drop into child droplets which provide ideal conditions for a complete combustion. Several authors have investigated the influence of several parameters on the micro-explosion phenomena as heating temperature [11-13], quantity of surfactant or emulsifier [11-15], or size distribution of water [11-19]. These papers outlined the major role played by the coalescence of dispersed droplets on micro-explosion occurrence, however, without providing quantitative correlation. In a previous work [11], Moussa et al. carried out a parametrical study where micro-explosion occurrence was investigated for different conditions, as a function of the Ohnesorge number (Oh). A particular attention is paid to ensure the same initial parameters for all the drops tested (same size, composition...). Indeed, the emulsion elaboration lasts long enough (15 to 20 minutes) to ensure a steady and homogenous mixture. Micro-explosion occurrence is defined as the ratio between the number of micro-explosion and the number of drops tested while Oh relates the viscous forces to inertial and surface tension forces and its expression is:



(1)

Figure 2: Influence of Oh number on the micro-explosion occurrence [11]

Where μc is the continuous phase viscosity, $\gamma_{W/O}$ is the interface tension between water and oil, ρ_w the water density and D the water droplet average size. From the Fig.2, we can see that micro-explosion is favored for low Oh emulsions (low viscosity, high interface tension and size). This can be explained by an enhancement of the water dispersed droplets motion through the oil phase, which leads to their coalescence. However, High Oh emulsions prevent the water droplets motion and cause a decrease of the micro-explosion rate.

More recently, Moussa et al. [20] conducted an experimental campaign, aiming to identify the drop whose phase change triggers the atomization (called trigger drop), and try to correlate its

properties (size and position) to the occurrence of micro-explosion. Size measurements were performed using Laser Induced Fluorescence (LIF) based techniques with a tracer soluble only in water. This configuration creates the visual contrast needed to identify water droplets from oil phase. Results showed that micro-explosion does not rely on the size and position of the trigger droplets, since it can happen when the trigger droplet is in the center of the emulsion drop or near the interface air/emulsion and is not dependent on its size.

1.2 LIF technique for temperature measurements

Laser Induced Fluorescence is an optical, non-intrusive technique allowing the measurements of different properties like concentration of species, temperature and size distributions. It involves the dissolution of a chosen tracer in the gas or liquid to be studied. Each tracer is characterized by its absorption and emission spectrum [21].

The intensity of the emission light I_f depends on several parameters [22]:

$$I_f = \int_{\lambda - \Delta \lambda}^{\lambda + \Delta \lambda} K_{opt} K_{spec} C_t I_0 v e^{s/T} d\lambda = K_{opt} K_{spec} C_t I_0 v e^{A/T^2 + B/T}$$
(2)

Where K_{opt} and K_{spec} are optical constants, C_t the tracer concentration, I_0 the intensity of excitation light, T the temperature of the medium and v the collection volume. A and B are constants related to the temperature.

For the configurations where only one detection wavelength is used, temperature can be measured thanks to Eq.3, where A and B are previously computed thanks to a calibration process:

$$ln\left(\frac{I_{f}}{I_{f0}}\right) = B\left(\frac{1}{T} - \frac{1}{T_{0}}\right) + A\left(\frac{1}{T^{2}} - \frac{1}{T_{0}^{2}}\right)$$
(3)

Where T_0 is the reference temperature, generally room temperature.

However, this method presents few drawbacks when applied to the temperature measurements of emulsion drops. All the parameters presented (except temperature) in the Eq.2 must remain constant in order to measure the intensity evolution generated only by temperature changes. This is hardly the case when it comes to the heating of emulsified fuel droplets. Indeed, for high temperature environments, it is highly probable that the tracer concentration C_t increases because of droplets evaporation. Moreover it is difficult to quantify the collection volume when coalescence, evaporation and fragmentation of droplets may occur.

In 2001, Lavieille et al. [23] improved the temperature measurement using LIF by introducing the two colors LIF (2CLIF). This method relies on the measurement of fluorescence emission intensity on two different wavelengths λ_1 and λ_2 and was used for the temperature measurements of mono-disperse ethanol drops jet and evaporating ethanol drops. Considering two different wavelengths we obtain two different fluorescence intensities I_{f1} and I_{f2} :

$$I_{f1} = K_{opt,1} K_{spec,1} C_t I_0 v e^{A_1/T^2 + B_1/T}$$
(4)

$$I_{f2} = K_{opt,2} K_{spec,2} C_t I_0 v e^{A_2/T^2 + B_2/T}$$
(5)

The fluorescence ratio R_f , defined as the intensity ratio of I_{f1} and I_{f2} can be expressed as:

$$R_f = \frac{K_{opt,1}}{K_{opt,2}} e^{(A_1 - A_2)/T^{2} + (B_1 - B_2)/T} = K e^{\alpha/T^{2} + \beta/T}$$
(6)

$$\alpha = A_1 - A_2 \tag{7}$$

$$\beta = B_1 - B_2 \tag{8}$$

Considering the fluorescence ratio R_f instead of the fluorescence intensity I_f solves the issue of the concentration dependence. Besides, the influence of the collection volume, incident light intensity are also eliminated by computing the fluorescence ratio on two different wavelengths.

2. Calibration of the 2CLIF method

The calibration of the 2CLIF technique is a critical step in measuring temperature of water dispersed droplets in oil. To this aim, it was studied a single demineralized water drop, previously mixed with Rhodamine B, and embedded in a silicone oil bath. This setting is close enough to the emulsion configuration and enables us to fix the water drop's position in a non-intrusive way.

2.1 Experimental set up and procedure

The Figure 3 and 4 represents the experimental setup used for the study of the fluorescence ratio R_f as a function of temperature.



Figure 3: Experimental design for the calibration of the LIF2C method



Figure 4: Top view of the experimental set-up

2.1.1 Heating procedure

The aim of the study is to measure the influence of the temperature on the evolution of the fluorescence ratio R_f . The heating device is a VMS-C4 (VWR) heat plate on which the cuvette relies. The temperature is measured thanks to a PT100 sensor which is connected to a proportional-integral-derivative (PID) controller, allowing the temperature to be measured and regulated.

The cuvette containing the different fluids is made of quartz with a square section of 10x10 mm² and a 45 mm height. Quartz is chosen for two main reasons. First, it offers an optimal transmittance for visible light (>95%), and moreover it is also known for its thermal resistance. The quartz cuvette contains an aluminum support, selected for the high thermal conductivity, on which the water drop is placed. Both the support and the drop are embedded in a silicone oil bath. Silicone oil was chosen as the continuous phase because of its high viscosity that inhibits the water drop displacement. Besides, silicone oil absorption (and excitation) is not concerned by the wavelength range we're interested in, and thus it will not affect the measurements. Thus, we should assume that changing tetradecane into silicone oil doesn't induce any bias.

The measurements are conducted when a stationary state is achieved. When the fixed temperature is achieved, we wait a few minutes for the temperature homogenization and then, we illuminate the water drop with a λ =532nm laser beam (Nd-YAG 2nd harmonic, Spectra Physics). The 5.5 Watts laser beam is transformed to a planar sheet, whose thickness is around 150 µm, by means of a pair of concave lenses. The fluorescence emitted light is then acquired and visualized thanks to the setup described in the next section.

2.1.2 Acquisition and optical systems

The light emitted by the fluorescent tracer inside the water drop is recorded by a Photron V4 Fastcam high speed camera mounted perpendicularly to the incident beam. It allows the capture of pictures up to 12000 fps with a maximum resolution of 1080x1024 pixels. However, in this paper, the frequency acquisition is fixed to 5000 fps with an exposure time of 0.125 ms. The high speed camera is equipped with a "Gemini" optical splitter device (Hamamatsu) (Fig.5).



Figure 5: Scheme of the optical splitter device

The Figure 5 illustrates the optical path of the incident light to the camera sensor. It's a three steps process. First the incident light is splitted into two beams by the means of a dichroic mirror with a defined separation wavelength. The beams go through two band pass filters, defined by the two detection wavelengths λ_1 and λ_2 (550 and 600 nm for Fluorescein, 600 and 650 nm for Rhodamine B) and are focused on the camera sensor thanks an optical lens. Using this device enables the visualization of the same event at two different wavelengths, as we can see in the Figure 6.



Figure 6: A water drop mixed with Rhodamine B visualized at λ =600 and λ =650 nm

2.1.3 Image treatment process

The raw images recorded by the acquisition system are treated with the Image Processing Toolbox of Matlab. The Fig.7 highlights the different steps of the process. First, the two similar size images $(m \times n)$ are extracted. They undergo some translation/rotation operations in order to accurately overlay one picture on the other. Each pixel of the resulting image corresponds to the intensity ratio of the two initial pictures (Eq.9):

$$ratio(i,j) = \frac{intensity \,\lambda_1(i,j)}{intensity \,\lambda_2(i,j)} \tag{9}$$



Figure 7: Image treatment process

2.1.4 Studied configurations

The key point that should be controlled is the amplitude of the fluorescent light signal. Indeed, it should be high enough to be detectable by the sensor, but always below the saturation threshold. Several parameters govern the emitted light intensity, as described in the Eq. 2. Thus, a dedicated study was carried out to define the most appropriate configuration of the experimental system.

The first critical aspect we are called upon to take is the choice of the tracer because of some constraints.

-The tracer needs to be soluble in water but not in oil. This a necessary condition to create the contrast needed for the visualization of the water embedded droplet.

-The tracer has to be excitable by a 532 nm laser sheet.

-The fluorescent light emitted by the tracer needs to be temperature-sensitive, between 20 and 150°C and its amplitude suitable with the actual acquisition system.

The two tracers selected are Fluorescein and Rhodamine B. The fluorescence was detected at 550 and 600 nm for Fluorescein, and 600 and 650 nm for Rhodamine B. In the Table 1, are presented the experimental conditions studied. As it was stated, different configurations are tested (Tab.1).

	Fluorescein		Rhodamine B		
λ (nm)	550	600	60	0	650
$C_t (mg/L)$	250	300	25	50	100
$I_0(W)$	5.5 & 7.5		5.5 & 7.5		

Table 1: Experimental conditions

2.2 Preliminary results

2.2.1 Tracer selection

The Figure 8 illustrates the evolution of the fluorescence ratio R_f as a function of temperature for both Rhodamine B (a) and Fluorescein (b).



Figure 8: Fluorescence ratio Rf vs temperature for Rhodamine B (a) and Fluorescein (b)

As it is highlighted, R_f decreases with temperature for Rhodamine B with a ratio range between 1 and 3. The trend is different for Fluorescein. Indeed the ratio is increasing between 2 and 3. The increasing of the ratio means that the intensity itself is increasing with temperature, which may represent an advantage because of the quality enhancement of the images.

However, in the fluorescein case, the trend does not seem to be monotonous as two local maxima can be observed. This implies that the relation between R_f and temperature is non bijective and therefore the fluorescein cannot be selected. All of the following experiments and results are conducted using Rhodamine B.

2.2.2 Validation of the initial assumptions

As previously stated, different configurations are tested for the study of the influence of parameters like the tracer concentration (Fig.9 a) or the incident light power (Fig.9 b).



Figure 9: Influence of the Rhodamine B concentration (a) and the incident light power (b)

The most remarkable result emerging from the data in the Fig.9 is the non-dependency of R_f on both the concentration ratio and the incident light power, despite some variation caused by

experimental errors. These findings confirm and validate the assumptions made in the section 1.2.

2.3 Estimation of the coefficient

The fluorescence ratio R_f can be expressed as a function of the medium temperature (Eq.6). If we consider a reference ratio R_{f0} computed at $T = T_0$ (the room temperature is around 20°C), we obtain:

$$\ln\left(\frac{R_f}{R_{f0}}\right) = \alpha\left(\frac{1}{T^2} - \frac{1}{T_0^2}\right) + \beta\left(\frac{1}{T} - \frac{1}{T_0}\right) \tag{10}$$

A variable substitution can be operated, thus obtaining a second order polynomial.

$$Y = \ln\left(\frac{R_f}{R_{f0}}\right) \tag{11}$$

$$X = \left(\frac{1}{T} - \frac{1}{T_0}\right) \tag{12}$$

$$Y = \alpha X^2 + \left(\beta - \frac{2\alpha}{T_0}\right) X \tag{13}$$

Figure 10 (a) illustrates the evolution of R_f as a function of the temperature of a water droplet mixed with Rhodamine B. The tracer concentration is 100 mg/L and the incident light power is fixed at 5.5 Watts. After the application of the variable change (Eq.11-12), we can plot the evolution of Y as function of X.



Figure 10: Evolution of Rf as a function of temperature and estimation of the coefficients

Figure 10 (b) indicates that the second order polynomial is suitable to fit the experimental data, with a good accuracy determination coefficient better than 0.99. From the coefficients of the fit, one can compute the values of α , β and K and thus, by using the Eq.(6), to correlate the fluorescence ratio to the medium temperature In this specific case, $\alpha = -1.79 \times 10^6$, $\beta = 1.21 \times 10^4$ and $K = 3.03 \times 10^{-9}$.

This procedure is then applied to several droplets in identical experimental conditions. Six cases were selected because of their high determination coefficient (\mathbb{R}^2 >0.97). The Table 2 lists the values of the coefficient α , β and K computed for each case. The average values of these parameters were integrated into a "model correlation" that will be used for the temperature measurements. The Figure 11 shows the R_f evolution with temperature for all the cases selected and the model correlation.

Case	α	β
1	-2.0×10^{6}	1.3×10^{4}
2	-2.7×10^{6}	1.8×10^{4}
3	-9.5×10^{5}	1.4×10^{4}
4	-2.1×10^{6}	7.3×10^{3}
5	-1.8×10^{6}	1.0×10^{4}
6	-1.6×10^{6}	1.2×10^{4}
Theo. model	$-1.7 imes 10^{6}$	$1.3 imes 10^4$

Table 2: Calibration coefficient for all the cases selected

The next step consists of expressing temperature as a function of the ratio (Eq.14), by computing the inverse function of the Eq.6.



Figure 11: Evolution of R_f with temperature for all the cases selected

The presence of square root induces one condition. The term $\beta^2 + 4\alpha \ln(\frac{R_f}{K_{opt}})$ should be positive. Thus, there is a critical value of $R_{f,crit}$ above which the result (temperature) be will complex.

$$R_{f,crit} = K_{opt} e^{-\beta^2/4\alpha}$$
(15)

All the results presented are deduced from experiments conducted at atmospheric pressure. For this reason, the temperature could not exceed the boiling point of water, which is 100°C. The range studied is [20-100°C]. However, during the heating of water-in-oil emulsion droplet, temperature of the water dispersed droplet may achieve metastable levels. This means that the water temperature is higher than 100°C, which is hardly reproducible for calibration

experiments. For these specific cases (temperature $>100^{\circ}$ C), the temperature is computed by extrapolation of the theoretical model.

3. Validation of the measurement method

The aim of this section is to validate the 2CLIF method for temperature measurements. The 2CLIF results will be compared to thermocouple measurements during the heating of a W/O emulsion drop under Leidenfrost effect.

3.1 Experimental method

The experimental setup bears a close resemblance to the one used in a previous paper [20]. However, we adapted it for the temperature measurement which requires two detection wavelengths by using an optical splitter device.

The acquisition and visualization material is the same as the setup described in the Fig.5 and 6. Concerning the heating device, it consists of a customized heat plate and is composed on several elements (Fig.12). A 6 mm diameter aluminum drop holder is designed with a circular convex geometry in order to trap the emulsion drop, avoiding its lateral displacement and eventually its fall. A resistive wire is mounted around the drop holder and is connected to a power supply. Temperature of the plate is measured by means of a 80 μ m K-type thermocouple placed at the bottom of the drop holder, with an accuracy of ± 1.50 °C. The thermocouple is coupled with a proportional integral derivative controller for the temperature regulation.



Figure 12: Heat plate design. A: Aluminum support; B: Drop holder; C: Location of the thermocouple

In the present study, the emulsion studied is a demineralized water/n-tetradecane emulsion, with $5\%_{vol}$ of water. Demineralized water is favored because of its relative purity that enhances the recording quality. n-tetradecane, like silicone oil that can only be excited in the UV band. It will not affect the 2CLIF measurements based on visible light excitation and detection. SPAN 83 is added $(0.1\%_{vol})$ to the emulsion in order to lower the interface tension between the two phases and improve the system stability. Rhodamine B is the fluorescent tracer, with a concentration of 2.2 g/L. This concentration may seem high and different from the concentration used for the calibration process, but it is needed for the visualization. Indeed, in the case of Rhodamine B, the fluorescent light intensity decreases with the temperature. Therefore, a high concentration of tracer is needed in order to ensure the visualization at the final stages of the fuel drop lifetime. The emulsification is performed using a mechanical stirrer at 1500 rpm during 10 minutes at room temperature.

The recorded images undergo the treatment described in 2.1.3. As it is mentioned, each pixel of the resulting image corresponds to the fluorescence ratio computed. This ratio used for the temperature computation thanks to the Eq.14 (Fig.13).



Figure 13: Steps of the image treatment process

3.3 Qualitative validation

The Tab.3 shows the evolution of the temperature distribution with the emulsion drop. The emulsion contains $5\%_{vol}$ of demineralized water, $0.1\%_{vol}$ of surfactant (Span83) and n-teradecane. The micro-explosion happens at t=349 ms, and unfortunately the image quality at the moment of the atomization is reduced and therefore, it is not presented in the Tab.3.

Time (ms)	Processed image		Description
0	Thermocouple	150 °C	
		- 100 °C 50 °C	The emulsion drop is about to be put on the 500°C heat plate. We can see the thermocouple located few millimeters above the heat plate so that it can be embedded in the drop.
		0°C	



Table 3 : Qualitative analysis of the temperature evolution of water dispersed droplets

3.4 Quantitative validation

In this section, the 2CLIF results will be compared to the temperature measured by a 80 μ m K-type thermocouple located inside the emulsion drop. As we can see from the Fig.14, this process is applied to 4 different cases with identical initial conditions. The emulsion contains 5%_{vol} of demineralized water, 0.1%_{vol} of surfactant (Span83) and n-teradecane and the heat plate temperature is fixed at 500°C. For the 2CLIF method, the temperature is computed in a small area (10x10 μ m²) located close to the thermocouple. This configuration enables to compare both temperatures at nearly the same location, which is an important issue considering the temperature gradient within the emulsion drop.



Figure 14: Temperature evolution measured by both LIF2C method and thermocouple

The marked observations to emerge from the data comparison illustrated in the Fig.14 are:

-The maximum temperature achieved in the studied cases is around 100°C, which represents the boiling point of water at atmospheric pressure. The metastable state is inhibited by the presence of the thermocouple inside the emulsion drop.

-The 2CLIF results are not smoothed because we couldn't differentiate between noise oscillation and oscillations caused by physical phenomena that may happen inside the emulsion drop like phase change, puffing and mainly internal convection [24].

-Both methods seem to be in agreement whit each other as the trend is nearly the same.

The average error ΔT is lower than 5°C after 100 ms for all the studied cases.

4. Investigation of the micro-explosion onset

After the calibration and the validation of the 2CLIF method, the interest was laid on the temperature distribution of water dispersed droplets as well as its evolution during the lifetime of the emulsion drop. The emulsion contains $5\%_{vol}$ of water, $0.1\%_{vol}$ of surfactant and n-tetradecane. This composition is chosen because it offers a high rate of micro-explosion [20], and therefore, it's optimal for studying the atomization trigger. Regarding the random aspect of the atomization process, 58 drops were studied, resulting in 38 micro-explosions (65.5%) and 20 puffings (34.5%). As previously stated, a special effort is made in order to ensure that the 58 drops tested contains the same amount of water and that the size distribution is unchanged.

In this section, three aspects are considered. First, the temperature evolution of a single water dispersed droplet, then the temperature spatial distribution. Finally, we investigated the

influence of the trigger drop properties (size, temperature and position) on the quality of atomization.

4.1 Temperature evolution

The temperature evolution measurements rely on the processing of "n" successive images where the concerned water droplet is visible and identifiable. Internal convective motions within the emulsion drop can elicit the water droplet to move from the laser sheet and therefore to disappear. For this reason, the temperature evolution is computed for only few ms while the emulsion drop lifetime lies between 300 and 900 ms.

Two water drops (D1) and (D2) are studied. The first one has an ascending motion (Fig.15) and the second one a downward motion (Fig.16). Initially, the drop (D1) is located in the bottom part of the emulsion drop. It is important to remind that the heat source is located underneath the emulsion drop. The temperature gradient generates an ascending motion of the water droplet that affects its temperature evolution. As depicted by the Fig.16, the water droplet temperature decreases from 63 to around 45° C in 6 ms. For similar reasons, the temperature of droplet (D2) increases with time, gaining around 20° C in 5 ms (Fig.16).



Figure 15: Temperature evolution of a water droplet with a downward motion



Figure 16: Temperature evolution of a water droplet with an ascending motion

The presented evidences (Fig.15 and 16) suggest the existence of a "temperature cycle" that concerns the majority of the water droplets (when convective motion effects are higher than gravity's). The first phase is its temperature increasing resulting into the emergence of a density gradient (Boussinesq effect) as well as an interface tension gradient (Marangoni effect). As a result, it triggers the water drop ascending motion towards the top part of the emulsion drop, naturally cooler than the rest of the drop. Moreover, this water drop motion is also driven by the natural convection of the oil phase. From this moment the second phase is triggered: the temperature decreases noticeably, inducing the rise of its density. Thanks to gravity effects, the water droplet goes back near the heat source and another cycle is about to begin. It is hard to quantify the frequency of these "temperature cycles" due to the tridimensional aspect of the convective motions, but the order of magnitude ranges between 10-20 ms for smaller droplets up to 70-90 ms for bigger ones. It must be emphasized that our experimental set up allows us to visualize only the water droplets located in the laser sheet. Moreover, these convection motions could also be induced by mechanical means. Indeed, when the drop falls on the heat plate, it swells and gets deformed (this happens also after an intermediate puffing), which may induce a flow within the emulsion drop.

Convective motions are not the only phenomenon that influences the temperature evolutions. Indeed, between each cycle, the temperature of a given water droplet can vary because of coalescence events. The temperature of a water droplet created by the coalescence of two water droplets is the weighted average of these two droplets temperature (Fig.17). Coalescence permits the generation of bigger droplets, which reinforce the impact of gravity, inertia and drag force and reduces the internal convective motions. One coalesced water drop tends to stay next to the heat source, which may lead to faster temperature rise.



Figure 17: Temperature of water droplets before and after coalescence

After each temperature cycle, the temperature level increases. When it's getting close to the saturation temperature ($100^{\circ}C$ for the water), two scenarios are possible: The droplet can undergo additional cycles, its temperature can achieve metastable levels and its phase change may induce a micro-explosion. The other option consists in a phase change at low temperature ($100^{\circ}C$) which may generate a puffing. This point will be discussed further.

4.2 Temperature distribution within the emulsion drop

The heating method (Leidenfrost effect in this case) implies the existence of a spatial temperature gradient within the emulsion drop. In this section, the evolution of the temperature gradient is investigated through the temperature measurements of different water drops at the same moment (Tab.4).

Time (ms)	Images	Comments
43.2	OT=49,8 °C T=41,9 °C T=46,8 °C T=46,8 °C T=46,8 °C T=41,4 °C T=41,4 °C	This image is taken a short time (2 ms) before touching the heat plate. The water droplets temperature is homogenous around 45°C with a deviation of 3°C (less than the accuracy of the method)

97.2	T=37,8 °C T=62,7 °C O T=45,8 °C T=40,8 °C	A small spatial temperature gradient appears within the emulsion drop. The average temperature of the water droplets located in the top part is 35°C in average while it increases to 50°C for the droplets located next to the heat source
184	T=64,8 °C O D=51,8 °C T=51,8 °C T=51,8 °C	The first effects of convection motion are noticeable since the temperature distribution tends to get more homogenous.
264.8	T=71,7 °C T=71,7 °C T=71,4 °C T=73,9 °C	Same observations can be made. One may notice that the temperature undergoes a significant rise (+11°C in average)
366.4	$T=85,3 \circ C \bigcirc T=92,4 \circ C \bigcirc T=9$	This image represents the last moments preceding the puffing. The temperatures are getting close to saturation point except for the large water drop in the right part. This specific drop underwent a double coalescence, which can explain its surprisingly low temperature

Table 4: Evolution of the temperature distribution

The emulsion drop studied in the Tab.4 underwent a puffing generated by the phase change of the water droplets whose temperature is 92.4°C (Fig.18)



Figure 18: Puffing of the emulsion drop studied in the Tab.4

It is interesting to notice that the temperature of the other water droplets is consistently lower than the trigger drop's. The atomization occurs before the temperature homogenization within the emulsion drop. If the temperature of the surrounding droplets is higher, the first phase change can lead to the phase change of these droplets leading to a more powerful break up. This point is discussed in the next section.

4.3 Occurrence of micro-explosion

This section is dedicated to the study of the atomization of emulsion drops, containing 5% vol of water, 0.1% of surfactant (SPAN 83) and n-tetradecane. As mentioned earlier, 58 drops are considered, resulting in 38 micro-explosions (65.5%) and 20 puffings (34.5%).

The average lifetime of the emulsion drop is 482.7 ms. From the Fig.19 it appears that the lifetime of the emulsion drop is influenced by the number of intermediate puffing that occurred. Intermediate puffing can be defined as a water droplet phase change without causing the atomization (even partial) of the emulsion drop. For lifetimes higher than 550 ms, micro-explosion is dominant over puffing.



Figure 19: Emulsion drops lifetime as a function of number of puffings occured

For each drop investigated, our intention is to identify the trigger drop and measure its properties at the moment of atomization (temperature, size and relative position). The identification of the trigger drop is the key point of this study, and three different configurations are possible:

-The trigger drop is not identifiable due to a low image quality at the moment of phase change or because it is not crossing the laser sheet. This concerns 29 drops among the 58 studied (50%).

-The trigger drop is identifiable but disappears from the vision field few ms before the phase change. Its properties are then measured at $t = t_{ato} - \Delta t$, where Δt is the time delay between the atomization and the last moment where the trigger drop is visible and lies between 0.2 et 12.4 ms. This configuration concerns 13 among the 29 cases where the trigger drop is identifiable (44.8%).

-The trigger drop is identifiable and visible at the moment of the phase change. In this case $\Delta t = 0.$ (55.2%).

Two aspects need some clarification before the results presentation:

-The temperatures measured are average temperatures within the water droplet. For larger drops, it seems highly probable that temperature gradients exist within the water droplet. Unfortunately, the resolution of the acquisition system is not suitable for measuring such spatial gradient with accuracy, since each pixel of the acquisition system computes an average value of ratio R_f that is converted to temperature. However, the temperature gradient ranges between 5°C/mm for smaller water drops up to 20°C/mm for larger drops.

-The size and position of the trigger drops are "normalized". At the moment of the atomization, the emulsion drop size is not always the same, from one case to another. This is a consequence of the evaporation rate and the intermediate puffing occurring during the emulsion drop lifetime. For this reason, size is considered as non-dimensional parameter R_t , defined by the Eq.16:

$$R_t = \frac{D_{trig}}{D_{emul}} \tag{16}$$

4.3.1 Micro-explosion

As outlined earlier, 29 trigger drops were identifiable and among them 22 leaded to a microexplosion (76%). The position, size, temperature of these trigger drops are illustrated in the Fig.20.



Figure 20: Temperature, size and position of trigger drops that lead to micro-explosion

As displayed in the Fig.20, most of the trigger drops are located in the bottom part of the emulsion (17 out of 22 or 77%). This can be explained by the density difference between dispersed and continuous phase (water and oil). However, smaller trigger drops can be located in the top part of the emulsion drop, mainly because of ascendant convective motions, that are dominant for smaller drops. The non-dimensional size R_t range lies between 7.8 and 33%, with an average value of 18%. Temperature of phase change varies between 93 and 129°C. One may observe that smaller trigger drops are the warmer. This could be attributed to a faster temperature homogenization within the smaller drops and a wider temperature distribution within the larger ones (Fig.21).



Figure 21: Temperature gradient within a water droplet

Two cases can be examined in greater details (a) and (b), representing the respectively the trigger drop with a phase change temperature of 129°C and 93°C.



Figure 22: Micro-explosion of the emulsion drop (a)

The case (a) is depicted in the Fig.22, The trigger drop is circled in red. The temperature phase change is around 130°C, which create a brutal metastable phase change leading to break-up of the emulsion drop. As we can see in the Fig.22, the emulsion drop totally disappears from the view field within 0.2 ms.

The case (b) is presented in the Fig.23. As it was mentioned earlier, the phase change temperature in this case is 93°C. It may seem illogical that the phase change temperature measured is 93°C, since the saturation point is 100°C.

Few points can explain this observation:

- The accuracy of this temperature measurement methods needs to be taken into account

- The trigger drop temperature is not measured at the exact moment of atomization. The measure is made 5.8 ms before the atomization. It's highly that temperature of the trigger drop increased, since it is located in the bottom part of the emulsion drop.

-As stated below, the temperature measured is an average temperature. And since we stated that there may be a temperature gradient within the trigger drop, the measured temperature may not be representative of the phase change temperature.

- In this case, the micro-explosion is triggered by the phase change of two different droplets (Fig.23). The first phase change occurs at T=93°C and triggers the phase change of a second droplet which temperature is 98.5°C. It appears the first phase change has created perturbations (with the swelling and deformation of the emulsion drop) that lead to the second phase change.



Figure 23: Micro-explosion of the emulsion drop (b)

Therefore, micro-explosion can occur even if the trigger droplet temperature has not reached metastable temperatures, provided a good temperature homogenization within the emulsion drop. This observation corroborates the findings obtained by Moussa et al. [11] (Section 1.2). Indeed, it was concluded that micro-explosion is enhanced for low Ohnesorge emulsions, because of the ability of the dispersed droplets to move within the continuous phase. In other words, internal convection motions result in the temperature homogenization where the water droplets are acting as energy vectors, but enhance also the coalescence rate of the water droplets, leading to the increase of their average size and size distribution.

4.3.2 Puffing

Puffing occurred for 7 times over the 29 cases where the trigger drop is identifiable. The properties of the trigger drops are depicted in the Fig.24.



Figure 24: Temperature, size and position of the trigger drops that lead to a puffing

In average, the phase change temperature is lower for puffings (92°C) than micro-explosion (106°C). The non-dimensional size is also lower (14% against 19% for micro-explosions). When it comes to the size and position of the trigger drops, the same observations than micro-explosion can be made. Two cases (c) and (d) are considered, with a size of respectively 10 and 33% (Fig 25 and 26).



Figure 25: Puffing of the emulsion drop (c)



Figure 26: Puffing of the emulsion drop (d)

As illustrated in the Fig.25 and 26, many water drops can be potential trigger droplets. For the case (c), the phase change temperature is very low (86°C) and the temperature of the

surroundings droplets is not sufficient to trigger a second phase change. For the case (d), the phase change temperature is around 92°C. It could have triggered a micro-explosion but the position of the trigger droplet is not optimal, and the temperature of the surrounding droplet is not close enough to the saturation point in order to trigger a second atomization.

4.3.3 Criterion for optimal atomization

Temperatures and sizes of trigger droplets are presented in the Fig.27 for both puffings and micro-explosion. According to the Fig.27, the smaller is the trigger drop, the higher should be the phase change temperature.

One exception is observed (points circled in red) near the virtual line separating microexplosions from puffings. For these two cases, the size and phase change temperature are similar, but one drop leads to puffing when the other undergoes a micro-explosion. For this kind of configurations, there are two possible explanations:

- The trigger drop position can be an important parameter, since a trigger drop located near the interface emulsion/air is more likely to generate a puffing.

- Even if the phase change temperature of the trigger drop is not high enough, temperature homogenization within the drop can lead to a micro-explosion by triggering "chain" phase changes. As discussed earlier, the temperature homogenization is permitted by internal convection motions



Figure 27: Size vs temperature of all the trigger drops

Concerning the influence of the trigger drop position, we have computed the enthalpy level of each drop ΔH_d (Eq.17) as a function of the distance between the center of the trigger drop and the center of the emulsion drop:

$$\Delta H_d = m_d c_p \left(T_{trig} - T_{ref} \right) \propto T_{trig} R^3 \tag{17}$$



Figure 28: Energy level of the trigger drop as a function of the distance to the emulsion drop center

The enthalpy level of a given drop is proportional to both its radius R and its phase change temperature T_{trig} . As illustrated in the Fig.28, the higher is the distance from the emulsion drop center, the higher should be the trigger enthalpy level in order to trigger an optimal atomization. The points (1) and (2) are characterized by a relatively high level of energy, however the phase change temperature is low (respectively 85 and 91°C). For the drop (3), a micro-explosion occurred with a phase change temperature of 112°C, which compensates the large distance to the emulsion drop center.

To sum up, temperature size and position of the trigger drop are the three parameters influencing the quality of an atomization (either puffing or micro-explosion). When one or two of these parameters is not optimal, they can be compensated by the influence of the third one. However, they can be ranked by order of importance. If the phase change temperature is high enough (115-130°C), micro-explosion occurs regardless of the size and position of the trigger drop. For lower temperatures (100-115°C), micro-explosion can happen if the trigger drop's size is high enough to provide a sufficient enthalpy level, or if the temperature is homogenized within the emulsion drop. For temperatures between 90-100°C, position of the trigger drop can enhance micro-explosion over puffing.

Conclusion

To investigate micro-explosion of emulsion drops, the study first presents the design and set up of a non-intrusive temperature measurement method of the inside water droplets, based on fluorescence and called LIF2C. This method is well suited for the visualization of processes like the atomization of an emulsified fuel droplet under Leidenfrost effect. Indeed, thanks to this method, the quantification of the tracer concentration, collection volume and incident light intensity is no longer needed.

The first calibration step is crucial. The tracer type and concentration is chosen, as well as the two detection wavelengths. In our case, Rhodamine B is selected over the other tracers: first, its high solubility in water and negligible in oil, the temperature dependence of the fluorescence light intensity emitted, and its compliance with the incident light wavelength. First measurements showed appreciable results both on the quantitative and qualitative aspects, even

though they can be improved: The LIF2C calibration process is conducted for temperatures between 20 and 100°C, while some water drops can achieve metastable states.

Concerning micro-explosion occurrence, the influence of the temperature, size and position of the trigger drop were investigated. We also observed the importance of internal convective motions: Inside water droplet are frequently observed changing their temperature rapidly, while moving from the hotter bottom to the colder top of the emulsion drop undergoing Leidenfrost effect.

Successful micro-explosion is triggered in most cases by water droplets reaching the highest temperature, and the highest enthalpy depending on its distance to the center of the emulsion drop. To sum up, temperature size and position of the trigger drop are the three parameters influencing the quality of an atomization (either puffing or micro-explosion). When one or two of these parameters is not optimal, they can be compensated by the influence of the third one.

Future work should focus on the LIF 2C calibration at higher temperatures, in a high-pressure cavity. Increasing the pressure should increase the boiling point of water, and therefore, it should achieve temperatures higher than 100°C, while it is still liquid. Another focal point is the temperature evolution measurement of the oil phase that can be conducted using Laser Induced Phosphorescence methods. This information may enlighten us on the heating kinetics of the two phases as well as the temperature difference between them.

Acknowledgment:

The authors thank the Region Pays de la Loire (Chaire Connect Talent ODE) for the financial support.

Nomenclature:

Symbol	Description	Unit (SI)
γ	Interface tension	$N.m^{-1}$
λ	Wavelength	m
μ	Dynamic Viscosity	Pa.s
ρ	Density	kg.m ⁻³
α, β	Calibration constants	
A, B, K	Calibration constants	
C_p	Heat Capacity	J/kg.K
$\dot{C_t}$	Tracer concentration	mol/L
D	Diameter	m
HLB	Hydrophilic-lipophilic balance	
Н	Enthalpy	J
I_0	Laser Intensity	W
K _{opt}	Optical constant	
K _{spec}	Spectroscopic constant	
LIF2C	Two Colors Laser Induced Fluorescence	

Laser Induced Fluorescence	
Mass	kg
Ohnesorge number	
Particle Matter	
Radius	m
Fluorescence ratio	
Size ratio	
Time	S
Temperature	°C
Trigger drop	
Collection volume	m ³
Water-in-Oil	
	Laser Induced Fluorescence Mass Ohnesorge number Particle Matter Radius Fluorescence ratio Size ratio Time Temperature Trigger drop Collection volume Water-in-Oil

References:

[1] T.Barker, I.Bashmakov, L.Bernstein, J.Bogner, P.Bosch, R.Dave, O.Davidson, B.Fisher, M.Grubb, S.Gupta and K.Halsnæs, Summary for policymakers. In Climate change 2007: Mitigation of Climate Change: Contribution of Working Group III to the Fourth Assessment Report of the Intergovernmental Panel on Climate Change. Cambridge University Press, 2007

[2] S.Solomon, D.Qin, M.Manning, Z.Chen, M.Marquis and K.B.Averyt, Contribution of Working Group I to the Fourth Assessment Report of the Intergovernmental Panel on Climate Change, Cambridge, UK and New York, NY, USA, Cambridge University Press, 2007

[3] S.Vellaiyan, K.S.Amirthagadeswaran, The role of water-in-diesel emulsion and its additives on diesel engine performance and emission levels: A retrospective review, Alexendria Engineering Journal 55, 2463-2472, 2016

[4] L.Marchitto, R.Calabria, C.Tornatore, J.Bellettre, P.Massoli, A.Montillet, G.Valentino, Optical investigations in a CI engine fueled with water in diesel emulsion produced through microchannels, Experimental Thermal and Fluid Science 95, 96-103, 2018

[5] A.M.Ithnin, H.Noge, A.K.Hasannuddin, and W.Jazair, An overview of utilizing water-indiesel emulsion fuel in diesel engine and its potential research study, Journal of the Energy Institute, 87(4), 273-288, 2014

[6] Y.Khan, Z.A. Abdul Karim, F.Y.Hagos, A.R.A.Aziz, and I.M.Tan, Current trends in waterin-diesel emulsion as a fuel, The Scientific world journal, Article ID 527472, 2014

[7] P.Geng, E.Cao, Q.Tan, L.Wei, Effects of alternative fuels on the combustion characteristics and emission products from diesel engines: A review, Renewable and Sustainable Energy Reviews 71, 523-534, 2017

[8] Y.Hagos, O.M.Ali, R.Mamat, A.A.Abdullah, Effect of emulsification and blending on the oxygenation and substitution of diesel fuel for compression ignition engine, Renewable and Sustainable Energy Reviews 75, 1281-1294, 2017

[9] J.Shinjo, J. Xia, L.C.Ganippa, and A.Megaritis, Physics of puffing and micro-explosion of emulsion fuel droplets, Physics of Fluids 26(10), 103-302), 2014

[10] H.Watanabe, T.Harada, Y.Matsushita, H.Aoki and T.Miura. The characteristics of puffing of the carbonated emulsified fuels, International Journal of Heat and Mass Transfer 52, 3676-3684, 2009

[11] O.Moussa, D.Tarlet, P.Massoli and J.Bellettre, Parametric study of the micro-explosion occurrence of W/O emulsions, International Journal of Thermal Science 133, 90-97, 2018

[12] E.Mura, C.Josset, K.Loubar, G.Huchet and J.Bellettre, Effect of dispersed water droplet size in micro-explosion phenomenon for water in oil emulsion, Atomization and sprays 20(9), 791-799, 2010

[13] E.Mura, P.Massoli, C.Josset, K.Loubar and J.Bellettre, Study of the micro-explosion temperature of water in oil emulsion droplets during the Leidenfrost effect, Experimental Thermal and Fluid Science 43, 63-70, 2012

[14] V.Califano, R.Calabria and P.Massoli, Experimental evaluation of the effect of emulsion stability on micro-explosion phenomena for water-in-oil emulsions. Fuel 117(A), 87-94, 2014

[15] A.A.Rashid, I.M.Tan, Z.A.Abdul karim, and Y.M.Khan, Experimental study on influence of surfactant dosage on micro explosion occurrence in water in diesel emulsion. Applied Mechanics and Materials 819, 287-291, 2016

[16] H.Nam, An experimental investigation of micro-explosion in emulsified vegetablemethanol blend, Doctoral dissertation, University of Texas, 2012

[17] Y.M.Khan, A.A.Rashid, Z.A.Abdul karim, and I.M.Tan, Experimental investigation of micro-explosion occurrence in water in diesel emulsion droplets during the Leidenfrost effect, Energy & Fuels 28(11), 7079-708, 2014

[18] E.Mura, R.Calabria, V.Califano, P.Massoli, and J.Bellettre, Emulsion droplet microexplosion: Analysis of two experimental approaches. Experimental Thermal and Fluid Science 56, 1587-1603, 2014

[19] R.Calabria, F.Chiariello, and P.Massoli, Combustion fundamentals of pyrolysis oil based fuels, Experimental Thermal and Fluid Science 31(5), 413-420, 2007

[20] O.Moussa, D.Tarlet, P.Massoli and J.Bellettre, Insight of a W/O emulsion drop under Leidenfrost heating using LIF optical diagnostics, Atomization and Sprays 29(1), 1-17, 2019

[21] W.Chaze, O.Caballina, G.Castanet, F.Lemoine, The saturation of the fluorescence and its consequences for laser-induced fluorescence thermometry in liquid flows, Experiments in Fluids 57(4), 2016

[22] A.Delconte, Fluorescence induite par laser multibande appliquée à la mesure de température dans les milieux complexes, PhD Thesis, Institut National Polytechnique de Lorraine, 2009

[23] P.Lavieille, F.Lemoine, M.Lebouché, and G.Lavergne, Mesure de la température de gouttelettes en combustion par fluorescence induite par laser à deux couleurs: résultats préliminaires et perspectives. Comptes Rendus de l'Académie des Sciences-Series IIB-Mechanics, 329(8), 557-564, 2001

[24] D.V.Antonov, G.V.Kuznetsov, S. Y.Misyura and P.A.Strizhak, Temperature and convection velocities in two-component liquid droplet until micro-explosion. Experimental Thermal and Fluid Science, 109862, 2019