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Nucleation inception due to conductive heating in composite droplets: experimental observations and modelling

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Abstract

Nucleation inception of composite droplets at slow to intermediate heating rates is examined both experimentally and theoretically. In the present study, rapid conductive heating of composite droplets of water in Diesel fuel is presented. Composite droplets are formed in such a way that initially water forms the inner sub-droplet and the hydrocarbon fuel forms the shell around it. The composite droplet is placed on a hot substrate until the water reaches the nucleation temperature. At this stage either puffing or micro-explosion are expected to occur. As the composite droplet is heated, the inner water core often comes into contact with the substrate, leading to a more efficient heat transfer and therefore to even faster disintegration. The Onset of Nucleate Boiling temperature (T_{ONB}), also referred to as the nucleation temperature, pertains to quasi-steady processes, and differs from the saturation temperature (T_{sat}), which pertains to equilibrium processes.

The typical modes of development of the heating and phase change processes leading to puffing or to micro-explosion events are analyzed. First, the composite droplet is being heated due to its contact with the hot substrate. The temperature differences promote vigorous movement of the drop, sometimes to the point of its detachment from the surface, which causes significant temperature fluctuations. Using energy considerations, we came to the conclusion that nucleation occurs at the interface between the two liquids, and the bubble grows into the less volatile component, the Diesel fuel, as long as the mass fraction of the water doesn't exceed 50%. The location of nucleation affects the dynamics of growth, and the location and time to puffing/micro-explosion; therefore, it is an important parameter, that is often overlooked.

Keywords

Composite droplet, Heating, Onset of Nucleate Boiling (ONB), Puffing, Micro-Explosion.

Introduction

Bubble nucleation inception is a physical phenomenon that characterises the onset of a phase transition process in a liquid. The purpose of the research is to predict the nucleation temperature due to an isobaric process under slow to moderate heating rates for composite droplets.

When the time required for superheating the liquid is shorter than the time that is required for bubble shedding from the interface, T_{ONB} is higher than the equilibrium saturation temperature (T_{sat}). In that case, the liquid is considered metastable, which is a partially, or kinetically, stable system, as opposed to a thermodynamically stable system [1]. A deeper penetration into the metastable zone is possible for fast processes [2]. Knowledge of the rate at which a system evolves towards stable equilibrium is essential to any investigation of metastable liquids, because such systems can only be studied over intervals that are short compared to the characteristic time for the appearance of a new phase [3].

In our previous studies [4][5], focused on rapid pool boiling, we identified three regions for the nucleation temperature depending on the heating rate (fast, intermediate and slow). A thermodynamic model, taking into account kinetic issues, was developed for the fast region for various pure liquids at atmospheric pressure ($10^6<dT/dt<10^9$ K/s) [6]. The model requires knowledge of the saturation and spinodal temperatures in order to predict the nucleation temperature as a function of the heating rate. Using original and previously published experimental data for water, two correlations, valid in the range $0<dT/dt<10^6$ K/s for water at atmospheric pressure, were suggested in the follow-up study [4]. For the slow region, a heat transfer model was developed for water at atmospheric pressure under three different levels of subcooling [7]. Lack of experimental data was identified within the intermediate heating rate region.

When considering a single component droplet, nucleation is most likely to occur at pre-existing nucleation centers, either at the surface or somewhere within the droplet (impurities or colloids). The question arises where will nucleation occur in composite droplets.

Composite biofuel droplets have the potential to be a useful solution in the energy field due to depletion of fossil fuels [8][9], to reduce NO_x [10] and anthropogenic emissions [11], and for wastewater treatment [12].

The present study aims to examine the time evolution of a composite droplet placed on a hot substrate, and to identify the typical different stages in the heating process of such a composite droplet. We will focus on the slow and intermediate heating regions for a composite droplet of water and Diesel fuel such as hydrocarbon fuel.

Experimental technique

Two-component Water-in-Fuel (W/F) droplets comprising of water and Diesel fuel, where the hydrocarbon (HC) liquid initially forms a spherical shell around the inner water droplet, have been investigated. To generate a two-component droplet, we used two Thermo Scientific Finnpipette Novus dispensers with a volume increment of 0.01 μl [11]. The relative volume concentrations of the components in a combined droplet varied in the range of 10-90%. No special treatment was applied to the liquids.

The initial droplet radius (R_{d0}) was calculated during the analysis of video frames before heating. The initial radius of the inner water droplet was evaluated based on the volume of water injected via the pipette and ranged from 0.5 to 1.5 mm. Using a scale factor, we calculated four droplet diameters in various cross-sections and averaged them to obtain R_{d0}. Accounting for the camera resolution and the scale factor, R_{d0} was calculated with an accuracy of at least $\pm$0.001 mm [11].

The present paper is concerned with composite W/F droplets heated mainly by conduction, when they are placed on a heated substrate [13], as shown in **Figure 1**. The substrate temperature was measured and regulated by a National Instruments 9219 (DAQ) module, and two fast-response type thermocouples (platinum-platinum–rhodium thermocouple TC, 0‑1200 °C, with an accuracy of ±1 °C, and a response time of 0.1 s [14]). To explore the dynamic pattern of water droplet heating and to determine the main characteristics of its temperature field, we used the Planar Laser-Induced Fluorescence (PLIF) technique [16]. Rhodamine B was used as fluorophore which was involved in water. In typical combustible liquids (Diesel fuel, rapeseed oil, kerosene) Rhodamine B doesn’t dissolve [16]. The thermocouples were used for temperature calibration, while PLIF was used for measurements during the experiments. Video frames at 1000‑3000 frames per second, were processed using Phantom Camera Control software.

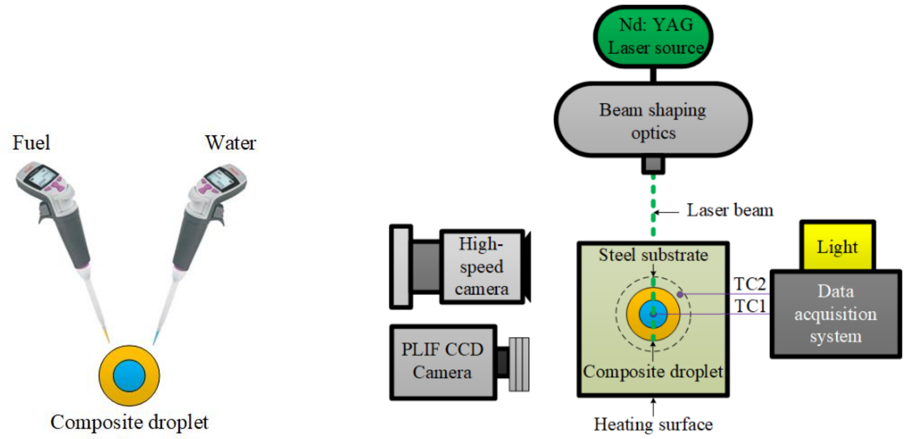


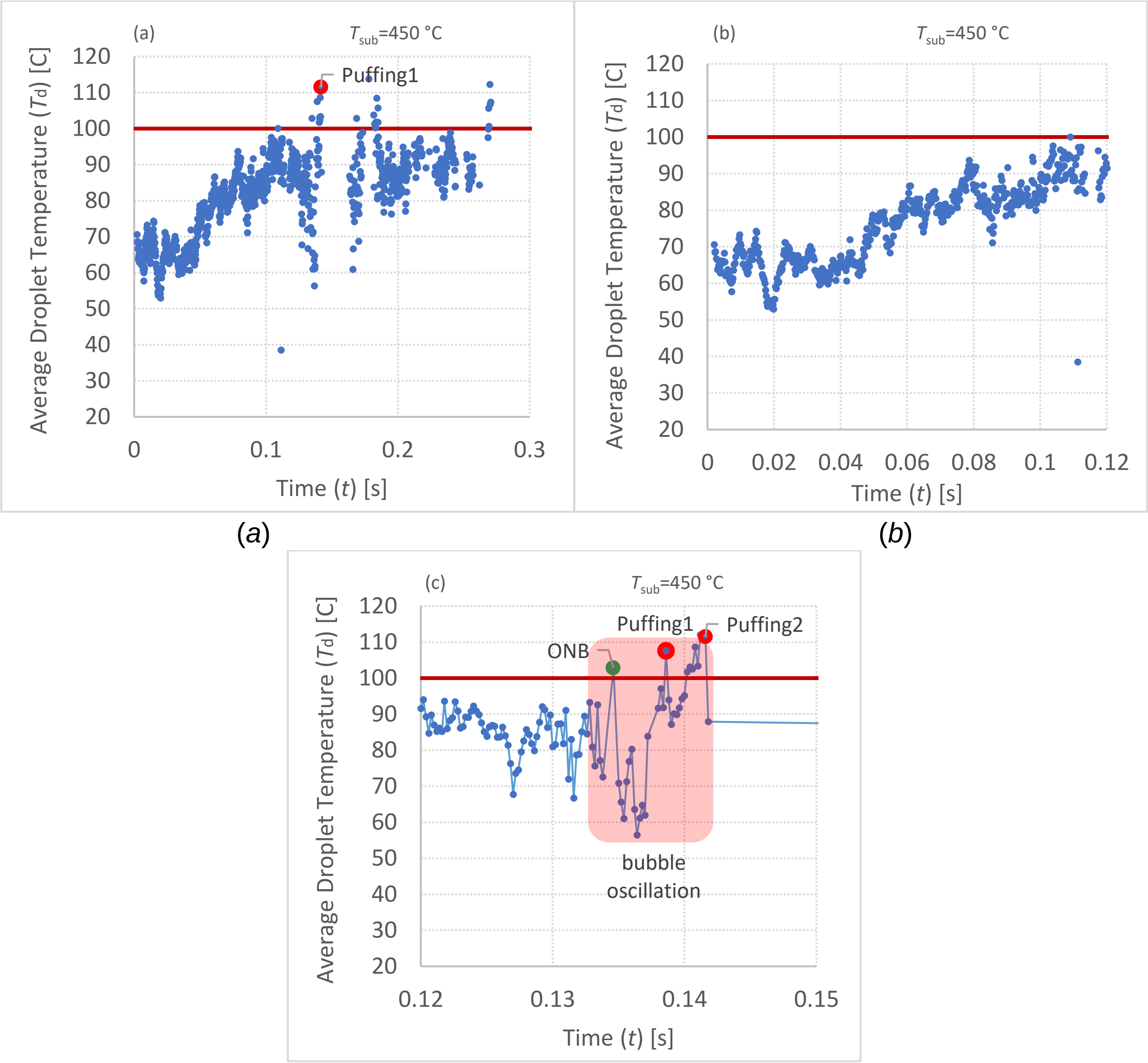
Figure 1. Schematic presentation of the conduction heating experiment for the composite droplet.

Results and Discussion

When a composite droplet is placed on a heated substrate, heat is conducted through the hydrocarbon liquid shell into the water sub-droplet. Since the saturation temperature of water at one atmosphere (100°C) is lower than that of most components of Diesel fuel (216.3°C, n-dodecane [17]), it is expected that the water will boil before the outer liquid hydrocarbons. Nucleation, however, occurs at higher temperatures, when the water sub-droplet interface is superheated. The degree of superheating can vary from a few degrees up to about 200°C, at the spinodal for water at atmospheric pressure. The actual superheating is influenced by many factors such as impurities and dissolved gasses within the liquids. In the cases where the inner droplet manages to touch the heating substrate, the morphology of the surface is of great significance. Since no special treatment was applied to the liquids, it is reasonable to assume that there are impurities and dissolved gasses within the liquids that serve as nucleation sites. These nucleation sites lower the energy barrier that is required for nucleation inception, thus the superheating degrees that were obtained in the current experiments varied between a few degrees and up to about 15 degrees.

The following figures present the time evolution of the interface temperature of the compound droplet placed on a hot substrate (T_{sub}=450°C). The interface water temperature at the interface, measured by PLIF (Rhodamine B was dissolved in water), increases over time. During the initial period (~0.04 s, **Figure 2**(a)) the oscillations may originate from the movement of the droplet on the substrate, and sometimes to the point of detachment of the droplet from the surface, as shown in videos. Subsequently (from ~0.04 s to ~0.12 s, **Figure 2**(b)) there is an overall monotonic increase in the temperature. Around t=0.126 s large oscillations commence. It is possible that these oscillations represent the oscillations of sub-critical vapour nuclei that form inside the droplet and collapse due to insufficient superheating of the droplet. This might cause oscillatory expansion and contraction of the droplet, until the degree of superheating of the droplet becomes sufficient for an ONB to occur at t~0.135 s. The oscillations due to the critical nuclei that can now grow become more violent. When the oscillations reach a critical size, they push the inner sub-droplet towards the surface of the droplet, thus either puffing (vapour erupts through the droplet surface) or micro-explosion (a large part of the droplet bursts) occurs. Since the droplet does not disintegrate completely at the first puffing event (t~0.139 s), multiple puffing events may follow; some may even be more violent than the first (e.g., t~0.142 s).

It is important to note that the onset of nucleation occurs at higher values than the normal saturation temperature for water at one atmosphere (T_{ONB}>100°C). In the current set of experiments, the degree of superheating reached up to 10°C. This was achieved due to the relatively high heating rate (~$2.5\ 10^4$ K/s). Yet, the degree of superheating for water at these heating rates can reach 25°C [4]. The low degree of superheating may stem from the fact that no special treatment was given to the liquids that composed the droplet, thus there were enough nucleation sites to promote early nucleation. The required size of these "ready-centres" has yet to be investigated.



(*a*) (*b*)

(*c*)

Figure 2. Typical experimental results for 10%vol of water – Average droplet temperature (T_d) vs. time for substrate temperature T_{sub}=450°C. The red dots indicate the puffing events. (*a*) Full scale experiment. (*b*) Initial period. (*c*) Towards the 1st puffing event: ONB, bubble oscillation and puffing (1st & 2nd).

The relative volume fraction of water in the above experiment was 10%. The mean droplet initial radius was R_{d0}=1.1 mm.

When the composite droplet is comprised of immiscible liquid components then depending on the surface tension of both liquids and on the interfacial tension, nucleation and bubble growth may occur either within either of the liquid phases or at the interface [18].

It is very difficult to accurately measure the surface tension at the interfacial region of two liquids. One of the methods for evaluating surface tension in mixtures was suggested by McGillis and Carey and is a simple weighted average (Section 2.4 in [19]). Although this approach should be used with caution, as it does not work for mixtures where one of the components serves as a surface-active-agent, they found it to be adequate for various mixtures of water such as water/ethylene-glycol. We have confirmed that it is adequate for water/freon-E9, by applying Jarvis et al. [18] expressions (see below) and comparing the results with the experimental results in their paper. Using this approach, we obtain in the case of water-Diesel fuel droplets:

$$\sigma_i = Y_{V,l,w}\, \sigma_w + Y_{V,l,Diesel}\, \sigma_{Diesel} \tag{1}$$

$$Y_{V,l,w} = \frac{Y_w v_{l,w}}{Y_w v_{l,w} + (1 - Y_w) v_{l,Diesel}} \tag{2}$$

$$Y_{V,l,Diesel} = \frac{(1 - Y_w) v_{l,Diesel}}{Y_w v_{l,w} + (1 - Y_w) v_{l,Diesel}} \tag{3}$$

where $Y_{\mathrm{V,l},w}, Y_{\mathrm{V,l,Diesel}}$ are the liquid volume fractions of the water and Diesel fuel, respectively, while Y_w is the mass fraction of the water within the mixture. $v_{\mathrm{l},w}, v_{\mathrm{l,Diesel}}$ are the pure liquid specific volumes for water and Diesel, respectively.

Jarvis et al. [18] argued that, if a more volatile liquid spreads on a non-volatile liquid, then less work will be required to create a vapour bubble entirely within the more volatile liquid. For that to happen the interfacial surface tension should be higher than the difference between the surface tensions of the two pure liquids:

$$\sigma_i > |\sigma_w - \sigma_{Diesel}| \tag{4}$$

Less work will be required to create and blow the bubble into the non-volatile liquid when the surface tension of the volatile liquid is higher than the sum of the surface tension of the non-volatile liquid and that of the interface. In other words, the interfacial tension should be lower than the difference between the surface tensions of the volatile and the non-volatile liquid:

$$\sigma_i < \sigma_w - \sigma_{Diesel} \tag{5}$$

According to these criteria, we found that for a water/Diesel fuel mixture the inception of the bubble will occur at the interface for mixtures containing mass fractions between ~50% and ~70% of water for temperatures from 20°C to 90°C. In most cases, the concentration of the component with the lower surface tension (here, Diesel fuel) is higher at the mixture interface, so we can deduce that the concentration of Diesel fuel at the interface will be higher than 50%, and thus, the mass fraction of water at the interface will be lower than 50%. Moreover, since nucleation occurs at high temperatures, it can be deduced that the bubble blowing scenario is the more plausible nucleation mechanism for all cases in the present study.

Summary

The boiling and disintegration of composite water/Diesel fuel droplets was investigated. Droplets with various volume fractions of water were placed on a hot substrate. After a short period of time (<1 s) the droplets experienced multiple puffing and micro-explosion events. The time evolution of the heating and phase-change processes were elucidated.

The temperature difference between the droplet and the hot substrate leads to vigorous movement of the droplet and to significant temperature fluctuations. The rapid heating rate leads to onset of nucleation at temperatures that are higher than the saturation temperature. The degree of superheating is determined by the competition between the heating rate and the sizes of the nucleation sites that exist within the liquid.

The location of nucleation affects the dynamics of the growth and the time to puffing/micro-explosion; therefore, it is an important parameter. Considering the thermophysical properties of the liquids in the composite droplet and their mass fractions, it is concluded that nucleation occurs and develops at the interface between the two fluids via a bubble blowing mechanism.

Nomenclature

R	radius [m]
T	temperature [K]
t	time [s]
v	specific volume [m^3/kg]
Y_V	volume fraction
Y	mass fraction

Greek symbols

σ	surface tension [N/m]

Subscripts

Diesel	Diesel fuel
d	droplet
i	interface
l	liquid
ONB	Onset of Nucleate Boiling
sub	substrate
w	water
0	initial

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