

Analysis of sub-processes of spreading and receding of sessile droplets on a hot substrate

Aliasghar Hajhariri¹, Florian Schulz^{*1}, Finn Duill¹, F. Beyrau¹

¹Lehrstuhl für Technische Thermodynamik, Otto-von-Guericke-Universität Magdeburg, Germany

*Corresponding author email: Florian.Schulz@ovgu.de

Abstract

In many technical applications, such as combustion engines, the investigation of the formation of liquid films attracts a lot of attention. The production of soot particles may be directly related to the unburnt liquid fuel film on top of the piston. Understanding the physics behind the simultaneous evaporation and receding of the liquid fuel film is not a simple task. However, this can lead us to model the behavior of film spreading/evaporation and ultimately reduce combustion by-products. To simplify the analysis of sub-processes of fuel film evaporation, studying a sessile droplet is proposed. Due to the complexity of the various phenomena that occur during spreading and receding of a multi-component droplet, conventional models do not take these processes into account. Therefore, we have investigated the time-dependent spreading/receding contact area and simultaneously described the occurring sub-processes that happen for different multi-component liquids on a heated surface. The dependence of temperature and contact area with respect to droplet lifetime are compared. Moreover, using schlieren imaging technique enables to reveal local evaporation during the whole process.

Keywords

Sessile droplet, Fuel evaporation, Liquid film, Mixture evaporation.

Introduction

Since the last decades, many researchers have worked on various hydrodynamic aspects of the liquid film layers. Nowadays, simulation and prediction of thin-film layer behavior is attracting attention. Many technical applications such as gasoline direct injection (GDI), ink-jet printers, and medical devices are affected by thin liquid film layer formation. For instance, inside the combustion chamber of a GDI, injected fuel can form a thin film layer that leads to fuel-rich zones. Consequently, these film layers lead to soot formation as a combustion by-product. Various numerical and experimental investigations have been conducted for one or multi-component liquids [1, 2]. The use of mixtures such as gasoline brings significantly more complexity to analysis. Therefore, scientists have utilized one-component fuels such as isooctane to simulate gasoline in several investigations. However, the spreading and receding behavior of real gasoline completely differs from that of isooctane. **Figure 1** reveals the sequences of the evaporation process of real gasoline and isooctane wall films over a hot substrate at 80°C. In order to predict exhaust emissions, the modeling of thin wall films is being intensively pursued [3]. This can only succeed, however, if corresponding experimental data is available for the validation of the simulation models. Besides, the spreading and receding of a film layer involves many simultaneous processes. Studies on thin liquid film layers are often performed using single sessile droplets. Most liquid sessile droplets are considered in the course of wetting where the evaporation process is completely neglected [4]. Analyzing the wetting process over a substrate has often been performed with pure liquids. One of the important properties that provide valuable information about the physics behind the processes of liquid film evaporation might be the contact angle, further interpreting the effects of various substrates [5, 6]. There is also a wide range of studies that have investigated the behavior of different one or multi-component

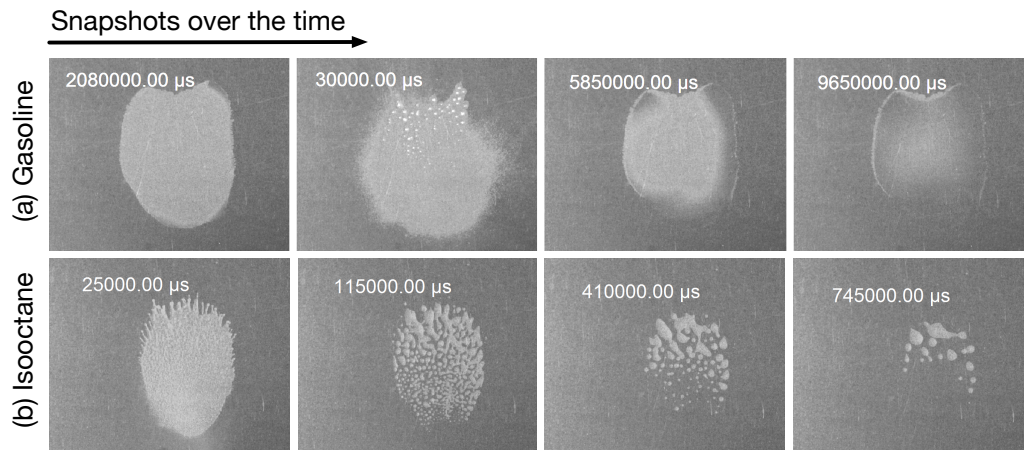


Figure 1. Film creation after injection at $p_{Rail} = 150 \text{ bar}$, $\vartheta_{wall} = 100^\circ\text{C}$ Gasoline (upper row) forms a flat film, and isooctane forms multiple single droplets (bottom row).

alkane droplets on various substrates. A great number of these investigations were conducted at room temperature and pressure. The main objective was to investigate radii, contact angle, and height of the droplets. D. Fardad & Ladommatos [7] investigated multi-component liquids over a wide range of substrate temperatures. Guéna et al. [8] investigated heptane-octane, hexane-octane, heptane-nonane binary mixtures on a silicon wafer. They recorded nominal radii of the contact area and evolution of structure over the droplet lifetime under ambient conditions. A large number of studies in the field of droplet spreading and receding processes during evaporation have been summarized in [2, 3, 9, 10], which also include theoretical considerations of complex processes in the case of sessile droplets. Although a considerable number of articles have attempted to introduce models that describe the behavior of a one-component film layer, a complementary model that can describe the behavior of multi-component mixtures is still missing. In order to distinguish the behavior of mixtures and real gasoline, moreover, to find the impact of the substrate temperature, we performed single droplet measurements on a hot substrate. For various temperatures, the present study gives access to qualitative wetted areas, and contact area of substrate temperatures during the spreading and receding process.

Material and methods

The visualization procedure of the spreading and receding of single droplets that form a very thin transparent layer is not a simple task. According to Schulz et al. [11] the fuel film residual formed on the cylinder wall and piston surface during the spray/wall interaction in homogeneously charged GDI engines is in the range of $5\mu\text{l}$. In the process, liquid droplets of equal volume are deposited over a hot substrate using a pipette. The influence of the wall surface temperature on the spreading and receding behavior of the droplet is investigated by varying the temperature range. Therefore, the initial surface temperature can be adjusted from 60°C to 140°C . The surface is heated in the analogous way to the setup in **Figure 2**. An Inconel 625 plate (nickel alloy) with a thickness of 0.1mm is heated using an electrical current, in which the joule heating provides a constant temperature. The arithmetic average roughness of the Inconel sheet is $0.24\mu\text{m}$ and the maximum roughness is $2.96\mu\text{m}$. The free surface energy is equal to 34.5mJ/m^2 . Recording the temperature of the droplet bottom was carried out by utilizing an infrared camera (IR8300 from InfraTec). Before the camera is being used, the backside of the substrate was painted with graphite spray to provide an emissivity equal to 0.89. This allowed the camera to be correctly calibrated so that monitoring of the temperature difference at the droplet contact surface from the backside is possible. The visualization of the liquid layer is carried out with the aid of camera (high-speed star 6 from LaVision). The goal of this setup is to record the droplet contact line, and area gradient during the spreading phase and follow-

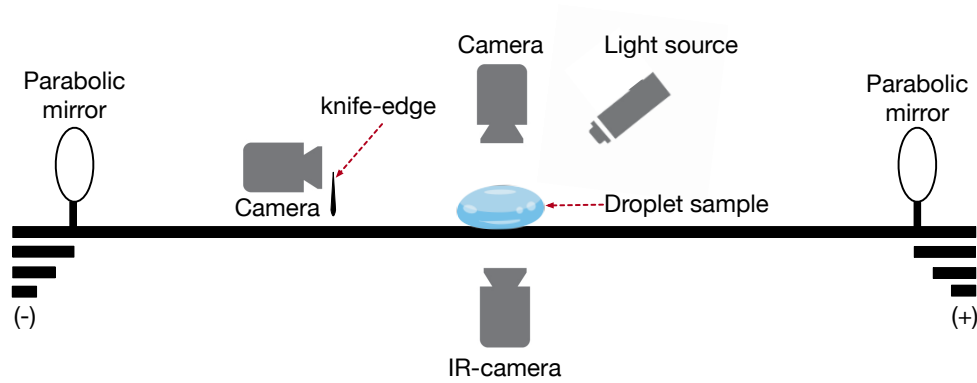


Figure 2. Schematic sketch of the experimental setup.

ing the receding phase. First, the light source (Dedocool-System from Dedotec) is adjusted to achieve a contrast between the transparent liquid layer and the reflective substrate (see **Figure 2**). For visualization, the high-speed camera with a 55mm lens is placed orthogonally above the droplet and records contact area of the droplet. Simultaneously, the temperature differences are captured with the aid of an IR-camera. This IR-camera is placed underneath the heated plate. Schlieren imaging enables us to evaluate the vapor phase over the droplet

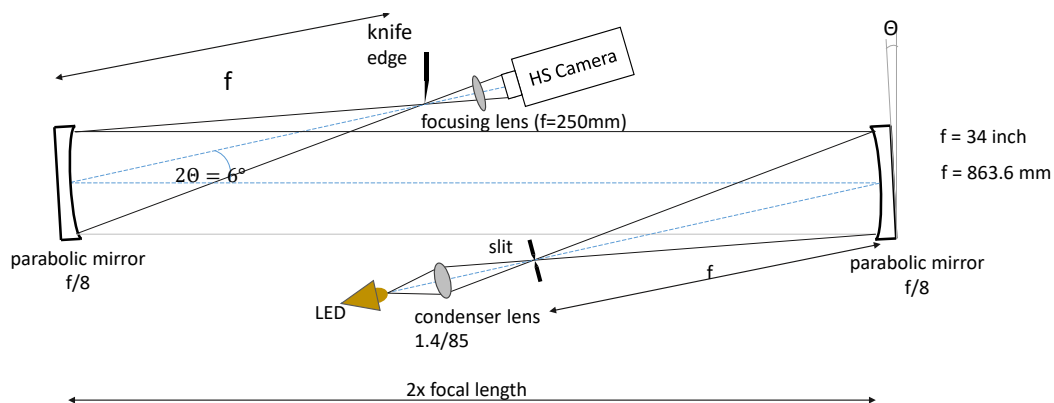


Figure 3. Z-type 2-mirror schlieren system

interface and provides qualitative information about local evaporation. Therefore, one more camera is used for capturing the schlieren images. Schlieren images together with the images from the top view of the droplet gives information about the locations where the high volatile component tends to evaporate. The schlieren setup depicts the changes in the refractive index, which is linearly proportional to the gas density. A z-type 2-mirror schlieren system was used in the measurements, shown in **Figure 3**, with two f/8 parabolic mirrors with a focal length of 864mm. For this, an LED was used as light source together with a 1.4/85 condenser lens and a source slit. The slit provides a homogeneous area of light that passes toward the first mirror. The reflected light passes the disturbed medium over the droplet where the evaporation takes place. The second mirror reflects the light toward a horizontally arranged knife-edge, further to a focus lens ($f=250$ mm) and, finally, to the camera. To investigate the spread and evaporation behavior of complex multi-component mixtures especially gasoline, bosch reference gasoline was utilized. Since real fuel behavior is difficult to model in CFD simulations, a simplified surrogate fuel which is called 3-component mixture [12] with a defined composition from hexane, isooctane and decane was tested as well. The 3-component mixture displayed in **Table 1**, illustrate the distillation curve of the applied gasoline.

Table 1. Characteristics of the tested multi-component fuels.

Notation	Description/composition
Gasoline	Bosch Reference gasoline (including 5 vol% ethanol) with 85% C, 13% H, 1.8% O and dist 10 vol% at 52.6°C, 50 vol% at 100.7°C, 90 vol% at 175.5°C
3-component mixture	45 vol % hexane, 35 vol % isooctane, 20 vol % decane

Results and discussion

To understand the strange behaviour of the multi-component mixtures such as real gasoline during the evaporation process, a sessile droplet of gasoline and 3-component mixture liquids have been analyzed. For the sake of comparison, the same procedure is applied to pure hexane, octane, and decane droplets. Concerning the impact of substrate temperature, the temperature varies between 60°C to 120°C. The evolution of the droplet area is recorded with a camera (see **Figure 4**). Interpreting the images enables us to distinguish between different phases during spreading and receding. With the aid of an IR-camera, the temperature at the center of the droplet where it contacts with solid substrate is achieved. The schlieren imaging technique depicts the local evaporation over droplet interface. The visualization procedure of little droplet evaporation over a hot substrate revealed various evaporation stages for multi-component mixtures that usually cannot be observed in the case of one-component droplets. The top view images depict the droplet evolution not only depends on droplet substances but also variables such as surface temperature make considerable differences. **Figure 5** and **Fig-**

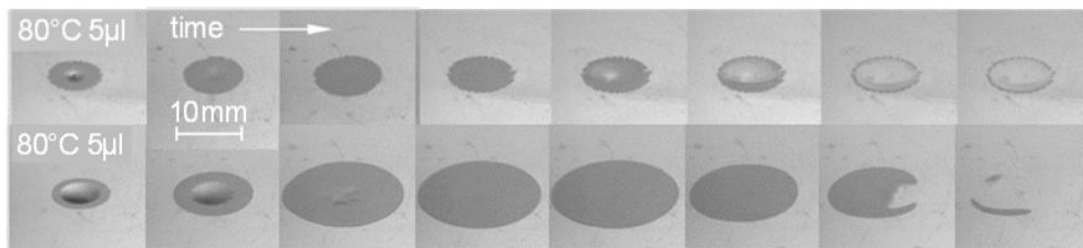


Figure 4. Gasoline (top) and 3-component (bottom) droplets spreading/receding during evaporation at $\vartheta_{wall,initial} = 80^\circ\text{C}$

ure 6 illustrate the time-dependent droplet contact areas, and the surface temperature of the center of the droplet that occupies the upper and lower part of the graph, respectively. In the following evaluation, the droplet areas are examined instead of the commonly used droplet radius. Due to dynamic changes during spreading or receding, it is not always possible to define a single droplet radius. **Figure 5** shows that the one-component droplets after deposition over the substrate start to spread slightly, however decane does not behave similarly. Although the spreading of one-component droplets is comparatively small, with increasing latent heat of evaporation (increasing the molecular chain length), the maximum area grows. For the sake of illustration, the decane graph is excluded on the **Figure 5(a)**. Generally, one-component droplets, e.g. hexane or octane, create a spherical cap that keeps its form until the end of the droplet lifetime. In other words, the contact angle remains in the range of contact angle hysteresis.

With respect to substrate constant energy, at the end of the droplet lifetime, the reduction of contact area continues. The temperature profile in **Figure 5(a)** and **Figure 5(b)** initially shows an intense temperature depletion that continues with a fast temperature incline after the first contact of liquid and solid surface. Since the cap structure provides enough height for creating a convective zone, the heat convection mechanism within the droplet homogenizes the temperature faster. Besides, from the droplet bottom toward its interface, there is a temperature

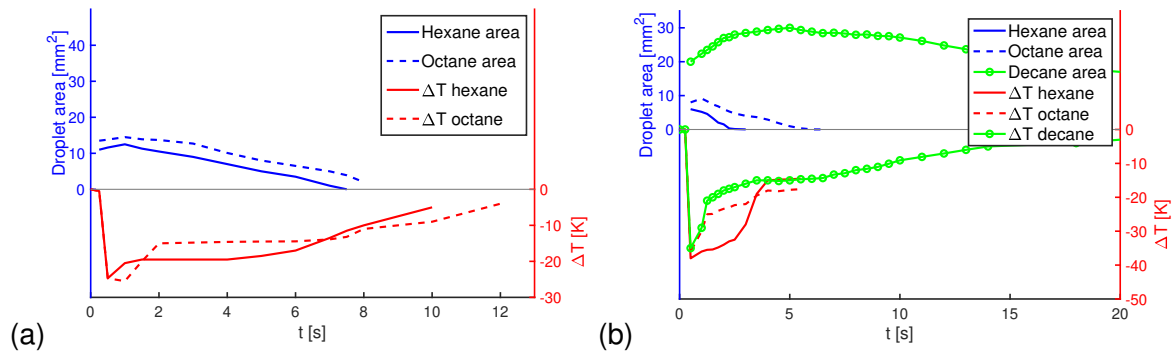


Figure 5. Area and temperature profiles of 5 μ droplets of hexane, octane, and decane droplets (a) at $\vartheta_{wall,initial} = 80^{\circ}\text{C}$ and (b) at $\vartheta_{wall,initial} = 100^{\circ}\text{C}$. Note that ΔT refers to the temperature difference of the droplet center on the substrate.

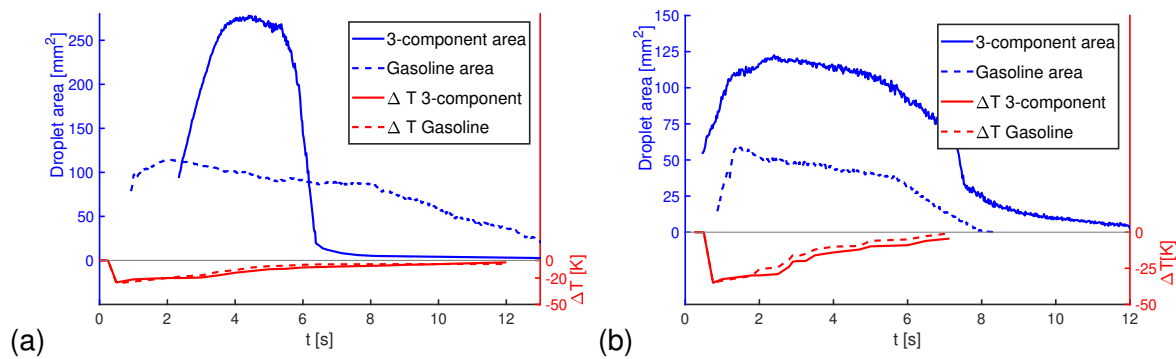


Figure 6. (a) and (b) indicate area and temperature profiles of 5 μ droplets of gasoline and 3-component droplets at $\vartheta_{wall,initial} = 80^{\circ}\text{C}$ and $\vartheta_{wall,initial} = 100^{\circ}\text{C}$, respectively. Note that ΔT refers to the temperature difference of the droplet center on the substrate.

gradient due to evaporation. However, a temperature gradient occurs when the droplet temperature is below the saturation temperature, e.g., for hexane the substrate temperature is already higher than saturation temperature, hence the temperature of droplets is assumed to be the same within the hexane droplet. After that, the temperature profile depicts a smoothly growing temperature region that contains around 40%-50% of the droplet lifetime. It may happen due to an equal rate of heat absorption by droplets from the substrate and the consumed heat for evaporation.

The phases and sub-processes occurring in multi-component droplets are briefly described in the following. Depending on the surface temperature of a hot substrate, various phenomena may happen. In contrast to one-component droplets, all multi-component drops form a flat structure as soon they are placed over a hot surface such that their contact angle are considerably smaller than pure drops and further extends with a meniscus. The meniscus is one of the reasons for the spreading of the droplet. However, hydrostatic forces due to droplet height cannot be neglected. Depending on the droplet and surface temperature, the the advancing meniscus forms fingers. Those finger-shape structures appear when the surface tension is low or the forces that drive movement can overcome the surface tension force [13]. This phenomenon has not happened for a 3-component mixture up to 100°C. In contrast to gasoline the finger structures might be seen for all temperatures. Extension of the fingers leads to the creation of some small droplets at the finger front. It happens for gasoline up to 120°C.

The maximum recorded areas of multi-component droplets from the top view are larger than the one-component liquids at the same substrate temperature except decane at 80°C (see **Figure 6**). For example, in **Figure 7** gasoline droplets spread more than octane, while the droplet lifetimes are comparable. After the evaporation of a large portion of the droplet liquid, only a thin liquid layer remains. The next stage is the receding phase that starts with a sudden area

reduction. It can be explained by a higher surface tension towards the droplet center which tends to reduce the contact area. For instance, **Figure 6** shows an intense area reduction of 3-component droplets after reaching maximum spread, whereas the gasoline area smoothly decreases. At the end of the droplet lifetime, the gradient of area reduction slows down, and temperature continues to increase until the droplet vanishes. An outer ring forms after spreading reaches its maximum for 3-component mixtures.

The second stage that comes after the spreading is the receding phase. This stage could divide into two various phases. The "dry out" and "contraction of the outer ring" are two processes that differ in this stage. Concerning the contact line position, if it is not pinned to the surface, similar to one-component droplets, the contact line retracts toward the centre. Another type of dry out occurs from the droplet's centre toward the contact line or vice versa. The second variant is the contraction of the outer torus that can happen when a portion of a liquid is trapped at the contact line. The amount of liquid at the torus does not evaporate completely leads to a reverse motion toward the center of the thin liquid layer. Hence, a new dome shape structure can be created or if there is still an old, dome the extra liquid will merge into it. This phenomenon has been observed for gasoline below 100°C. The temperature profile of multi-component drops, similar to one-component drops, shows an intense decline as soon as the first contact of the liquid and substrate, see **Figure 6(a)** and **Figure 6(b)**.

Later, the droplets start to gain heat, and the temperature grows to a certain amount. Considering a 3-component droplet, the droplet spreads farther than gasoline. The temperature profiles show when the substrate temperature increases, the droplets absorb more heat over small times. Therefore, initial temperature depletion becomes stronger. Besides, with increasing substrate temperature, the droplet lifetime decreases. After the multi-component mixture reached its maximum spread, residuals contain the low volatile component that evaporates slowly and stays much longer over the substrate. Hence, the droplet needs more thermal energy to compensate for evaporation.

Schlieren images depict a vapor profile over a droplet that forms a mushroom shape in **Fig-**

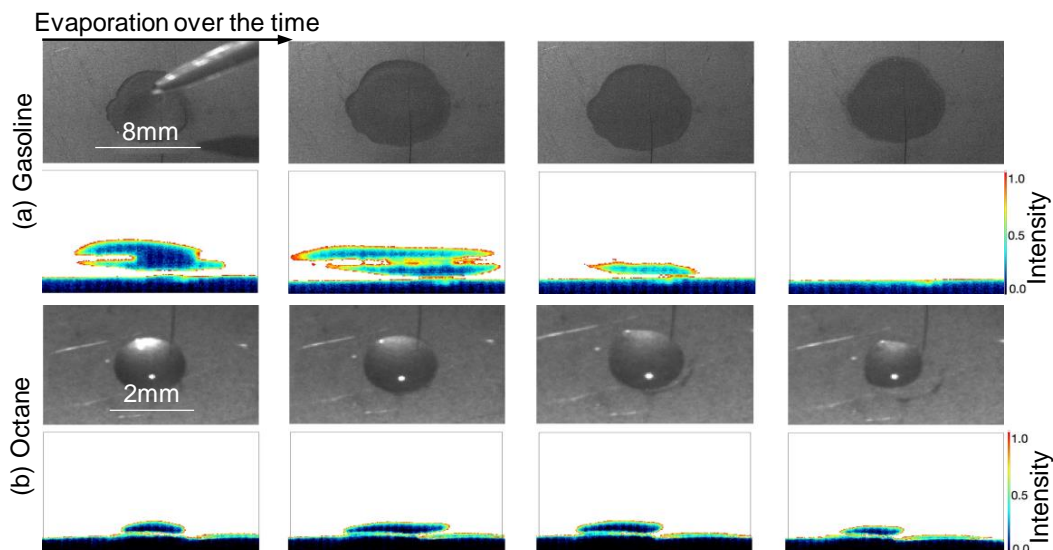


Figure 7. Top view images versus schlieren images during evaporation process for gasoline and octane droplets at $\vartheta_{wall,initial} = 80^{\circ}\text{C}$

Figure 7. The vapor profile points out convection and diffusion are driving forces for the vapor phase. Furthermore, the shape of the vapor profile in the vicinity of the contact line illustrates higher evaporative flux at the contact line which is in good agreement with [8, 11]. In the images of **Figure 7** where the schlieren images are illustrated in the second row of each liquid (gasoline and octane) there is a tiny gap (white line) close to the substrate between the surface and vapor phase that does not happen when the surface is at ambient temperature. Thus, the rea-

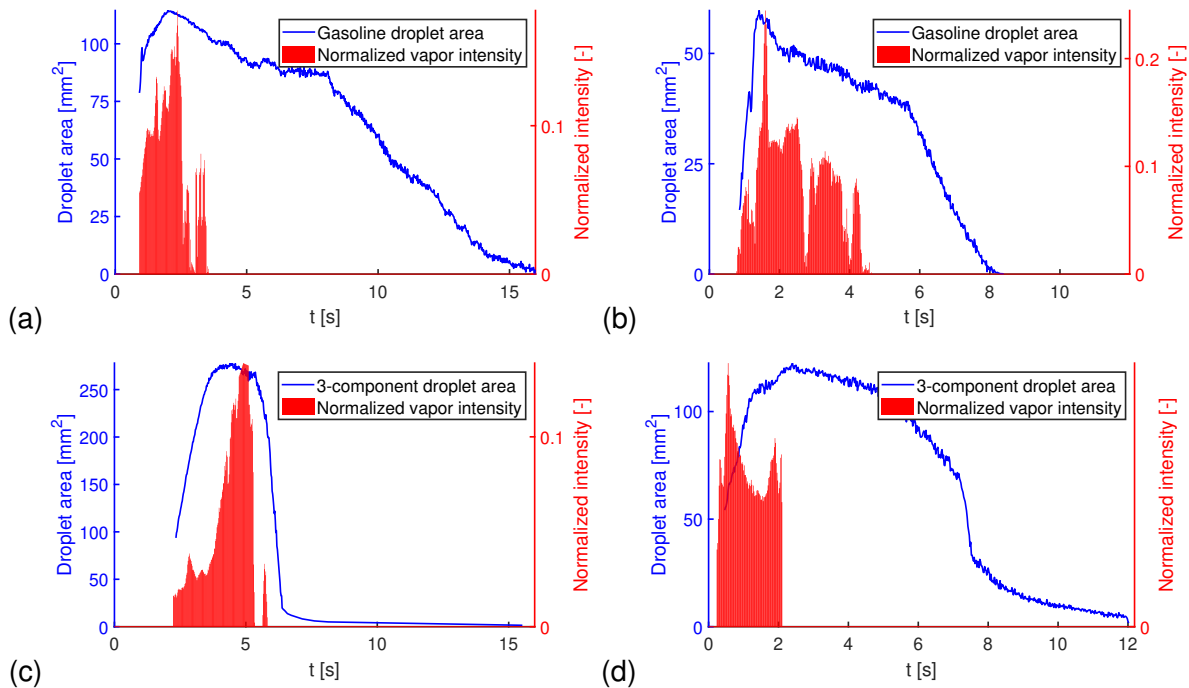


Figure 8. (a) Gasoline droplet area with normalized vapor intensity at $\vartheta_{wall,initial} = 80^{\circ}\text{C}$ (b) Gasoline droplet area with normalized vapor intensity at $\vartheta_{wall,initial} = 100^{\circ}\text{C}$ (c) 3-component droplet area with normalized vapor intensity at $\vartheta_{wall,initial} = 80^{\circ}\text{C}$ (d) 3-component droplet area with normalized vapor intensity at $\vartheta_{wall,initial} = 100^{\circ}\text{C}$

son for observing this gap is the density gradient of hot air over the substrate. The normalized intensity of the vapor over the liquid-vapor interface of each droplet is individually investigated. The light which passes through a denser medium is refracted, thus the darker region reveals higher vapor concentration. In the case of one-component droplets, the vapor cloud remains until the end of the droplet lifetime (see **Figure 7(b)**).

The intensity of vapor cloud of one-component liquids increases after droplet deposition and stays in an almost constant region. At the end of the droplet lifetime, the normalized intensity profile shows no considerable intensity value. It means the evaporative flux becomes slower. In other words, the convection near the hot substrate causes dilution and the density gradient decrease consequently. In contrast to one-component droplets, schlieren images of multi-component droplets show the vapor profile for only a part of the droplet lifetime that begins with droplet deposition and vanishes around the maximum spreading area. Initially, evaporation accelerates with increasing the droplet area until the maximum spreading is reached. The evaporation decelerates while the images from above show a very thin liquid film. Later, schlieren images reveal no intensity gradient that means the evaporation slows down with a significantly reduced amount of vapor above liquid film layer, in **Figures 8**. Therefore, it can be concluded that most of the high volatile components are preferably evaporated. It can also describe why the spreading area of multi-component is larger than one-component droplets. Due to the concentration gradient a large portion of the high volatile moves towards the contact line and motivates it to spread further. Herein, there is a good qualitative consistency between the area gradient that achieve from the top view in comparison with the vapor profile area from schlieren images. **Figures 8(a)** to **8(d)** depict that the normalized intensity grows where the spreading area reaches its maximum then it decreases.

Conclusions

In this study we could find increasing the surface temperature, decrease the evaporation duration. Octane spreads similar to mixtures that follow the gasoline trend. Concerning multi-

component droplets, 3-component spread larger than gasoline. Gasoline evaporate smoothly in contrary 3-component mixture experience a sharp area reduction. Thermography shows an intense temperature depletion that is driven at the initial contact of liquid with a hot substrate for all types of liquids. The heat energy that is radially conducted from the surface to the sample can provide the required energy of vaporization. Schlieren imaging shows a higher rate of evaporation at the contact line. This might be due to diffusion of the more volatile component toward the contact line. Furthermore, when the droplet is deposited over the substrate, differences in the substance concentration drive a Marangoni flow toward the contact line where the surface tension decrease and more evaporation observed. The receding process of multi-component droplets is quite individual. Partly, temperature influences the slop of area reduction. For instance, below 120°C the gasoline area is larger in comparison with the 3-component mixtures, and its lifetime is shorter, consequently. For a certain substance, increasing the droplet area follows by height reduction. Nevertheless, it increases the evaporation rate, it can change the heat transfer mechanism. At the maximum spreading of multi-component droplets, schlieren images show that the evaporation rate is not considerable, so it means the concentration of high volatile components reduces and less volatile components remain until it evaporates completely.

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