

Investigation of Fire Extinguishment Mechanisms at Microscopic Level

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Abstract

This study is aimed at investigating the microscopic behaviour of water droplets in fire extinguishment mechanisms. In particular, this study focuses on the qualitative and quantitative measurements of a water droplet interaction with smoke particles and solid fuel surfaces in a fire scenario. The main topics of this research are focused on fundamental small-scale processes by utilising various imaging techniques to study the static and dynamic behaviour of water droplet evaporation. The evaporation characteristics of water spray can be studied at a fundamental level by examining water droplet evaporation. Evaporation, vapour formation, liquid phase dynamics, and interactions of water droplets share similar processes with droplets in spray systems. Therefore, isolated droplet studies which involve energy transfer and mass transport are relevant to the complex environment of spray in fire extinguishment applications.

Various experimental techniques were developed and utilised in this present work. The development of digital imagining based on cameras and thermal detectors offered many advantages in the field of fire dynamics and combustion diagnostics. The non-intrusive nature of most imaging techniques enabled its utilisation in droplet studies. Image processing techniques were used in droplet evaporation, surface evaluation, and impact studies were outlined. Calibration of thermal cameras in different infrared bands was performed. Using synchronised technologies at the same time made it possible to acquire evaporation characteristics simultaneously and study interactions comprehensively.

The effect of suspended particles on evaporating droplets was investigated experimentally and numerically. Droplet evaporation was studied under different conditions along with different particle suspension methods. Quantitative measurements of evaporating rate showed that contaminated droplets had a higher rate of evaporation. The results were consistent with available research on nanoparticle effect in fluids. However, previous experimental research investigated the properties changes by volume mixing, while this study focused on surface contamination, which is most likely in a water spray fire suppression process. Furthermore, Water surface tension was evaluated using the pendant droplet method. Salt solutions were also investigated along with pure water droplets. Surface tension was found to be lower for surface-contaminated droplets. The droplet surface motion was found to changes in a way where clusters of particle networks move together around the surface.

A detailed analysis of the phenomenological, morphological, thermal, and topographical aspects of droplet interactions was presented. Significant processes of droplet spread evolution, the effect of droplet diameter, impact speed, and burning behaviour were explored and detailed using multiple imaging techniques. Droplet spread on surfaces created a high surface-to-volume ratio, leading to rapid evaporation. Immediate wetting of burning surfaces and subsequent bubbling is suggested to play a role in preventing the escape of volatiles to the reaction zone. Hence, a local mechanism based on the sealant effect of water liquid layer is proposed in addition to the cooling of fuel surface. Bubbles can act as a wall in the same way as the unfissured zone.

The investigated interaction between droplets and combustible surfaces shows the importance of microscopic behaviour in assessing the fate of combustion processes. This fundamental understanding paves the road for improved fire dynamics models and better technological approaches to extinguishment systems.

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Chapter 1 Introduction

1.1 Background

Over the years, several chemicals have been used to firefight and tackle fire. The look for efficient suppressant agents led to the use of halons. Since halons proved to be a major environmental and health challenge [1], [2], non-hazardous alternatives were sought. The exploration has led to promoting the use of water, which is non toxic, does not produce harmful by-products, and does not lead to detrimental effects on the materials.

Water is extensively used as the base fluid if not the only material in the systems of fire suppression. Because of its comparatively high specific heat capacity and latent heat of evaporation, water can essentially act as a heat sink in a fire, causing extinguishment by cooling a fire. In addition to the thermophysical properties of water, the method of delivery to the fuel and the involved dynamic interaction with smoke, flames, and fuel are similarly important when used as a suppressant in fire scenarios. Water is typically dispersed into small droplets and distributed to the burning area and objects as a fluid spray by means of water sprinklers or mist nozzles. Therefore, the effectiveness of water suppression action depends on the spray behaviour and interaction with fire. This action involves many extinguishment mechanisms, such as radiation shielding, oxygen displacement, gas phase cooling, and fuel cooling. Understanding the physical and chemical phenomena in each of these mechanisms enables better modelling of water spray behaviour and performance in a given fire scenario.

The current research on fire suppression focuses on sustainable extinguishment methods and the optimisation of conventional methods. Although there are promising new smart technologies in firefighting, water sprays are still the first choice and the most used in a variety of fire scenarios. Therefore, enhancing water efficiency in suppressing fire along with reducing the amounts of consumed water is crucial for demanding applications [3], [4], [5], one way to explore the interaction of water with fire is by examining droplet behaviour [6], [7], which enables specific studies into the mechanisms of fire extinguishment.

1.2 Importance of droplet interaction

Exploring droplet interactions in different fire scenarios is beneficial for the continuous improvements of the methodologies used in the area of fire suppression. More profound knowledge of water droplet evaporation and impact characteristics would provide comparative insights of spray systems [8]. The approach of scaling an isolated water droplet and spray systems would provide insights into the physical phenomena that can be reflected in numerical and modelling studies [9]. Moreover, observation of a single droplet behaviour and processes associated with evaporation are of importance in the prediction of many engineering tasks [10]. Therefore, isolation of droplet processes, which involve mass and energy transport, proved to be relevant to the highly complicated environment of fire interactions with suppressants in the form of sprays.

Several researchers have focused on the quantitative measurements of isolated droplet with the inclusion of deposited smoke particles including detailed dynamics throughout droplet evolution using imaging [11], [12]. Droplet impact on fuel surfaces [13], [14], droplet evaporation in high temperature environments [15], [16], and droplet flame interaction [17], [18], [19], [20] show to mainly influence physical phenomena, especially in reducing the resultant flame temperature, but it also points to evidence that water vapour interacts chemically in the flame. The approach of researching isolated droplets has been used to support the theoretical and analytical investigation of evaporation to include different physical and chemical processes. The evaporation process at the last stages was found to be controlled by the effect of surface tension caused by the droplet curvature [21]. Further research in the area will contribute to understanding the complex details that are hidden in the interaction of water droplets and fire.

Although there has been considerable research on the behaviour of single droplets and their interaction with various fire types, there is still a research gap in understanding the detailed interaction between individual water droplets and solid fuel surfaces. This gap is particularly relevant when considering the potential changes in droplet thermophysical properties due to particle deposition, as well as the impact of such changes on the overall fire extinguishment mechanisms. Existing research has typically assumed that droplets retain their base fluid thermophysical properties as they pass through smoke layers and flames. However, there is evidence to suggest that surface particle deposition can alter these properties and consequently affect the performance of water spray and droplets in fire suppression. This lack of data and understanding highlights the need for both experimental and analytical investigation into this phenomenon. A better understanding of these interactions can lead to improved fire suppression models and more effective firefighting strategies.

Moreover, most existing experiments on sprinkler and mist system performance have been conducted for specific fire scenarios and often at a large scale, making them expensive and less flexible. Small-scale experiments, on the other hand, can offer a unique approach to studying fire extinguishment, allowing for more flexibility and cost-effectiveness. Furthermore, there is limited research on the impingement of water droplets on burning solid fuel surfaces, such as wood. Investigating this process is crucial to understanding one of the primary aspects of fuel wetting by water-based systems.

The development of digital imagining and optical systems based on cameras offer many advantages in the field of combustion diagnostics. Imagining technology has also been used in other applications such as quality inspection, medical checks, and vibration analysis. The non-intrusive nature of most imaging techniques enables their utilisation in droplet studies. Furthermore, using more different technologies at the same time provides the ability to study interactions comprehensively.

1.3 Problem statement

Investigation of droplets' interaction with smoke, reactive phase and fuel beds assumes droplets retain their base fluid thermophysical properties. When water droplets pass through a smoke layer and flames, droplets have a potential for surface particle deposition. Performance parameters of water spray and droplets have not been investigated in this case. The lack of data and understanding is the motive and need to investigate this phenomenon experimentally and analytically. Once the research is established and validated, the improvement in modelling can be of great value to engineers and researchers. Moreover, the physical description can be used to develop a more accurate and reliable description of water spray behaviour in fire. Thus, the overall aim in this work is to explore the microscopic water droplet behaviour as used in fire extinguishment in order to broaden the fundamental understanding of extinguishment mechanisms in different applications.

Most experiments that study the performance of a specific sprinkler and mist system are run for a particular fire scenario. Furthermore, large-scale tests are expensive, and the options for designed fire are also wide, such as buildings, aircrafts, or storage facilities add to the complexity, let alone the cost. One can also argue that results from small-scale experiments are limited because the test conditions achieved [22]. However, small-scale experiments can offer a unique approach to studying fire extinguishment, allowing for more flexibility during experimentation [23].

Several experiments on smoke particles of lab-scale test fires and wildfires showed different distributions. Smoke particles which were examined immediately after production indicated a peak at 100 nm. While smoke transported by the air, which tends to agglomerate, exhibited peaks at larger sizes. The studies showed that smoke particles differ in their chemical constituents. The composition depends on the material that generated the smoke, carbon soot is the main prevalent agglomerate in wood combustion [24], [25], [26]. In addition, smoke contains metals such as iron, potassium, sodium, magnesium, and copper. Other materials such as phosphorus, silicon, and sulphates, can also be found. Due to the composition and size of smoke particles, it is expected that droplets with particles deposited on their surfaces have different surface behaviour. Rasid and Zhang [27] examined surface contamination in a fuel droplet and reported the impact of particles on thermophysical properties, concluding that surface contamination with soot significantly modified the characteristics of diesel droplet burning. They showed that evaporation and burning constants were lowered, and the particles affected flame stability critically. However, the focus was only on the burning behaviour of a fuel droplet without further experiments on evaporation as a separate process.

Repeatable experiments need reliable droplet generation. Droplet generators are important instruments used in many fields, including spray processes, printing, and surface impact studies [28], [29], [30]. Single droplet experiments are commonly conducted to study droplet formation, evolution, stability, evaporation, surface impact, and combustion [31]. Drop-on-demand techniques seem to offer accurate generation and offer high repeatability. However, challenges are faced when an attempt is made to utilise current devices in a variety of applications. Difficulties such as satellite droplets, shape distortion, bubbles and trajectory variation are common operation challenges. Recently, several new droplet generator constructions have been suggested for various reasons, including design simplification, improved control, fluctuation reduction and extreme working condition capability [32], [33], [34], [35], [36]. Therefore, a solution is needed to be applicable for combustion experiments and fire studies, and the device should demonstrate droplet generation for the intended utilisation in experimental work.

Various fire suppression mechanisms have been described as relevant to hydrocarbon fires, including fuel wetting and surface cooling [37]. Investigations regarding surface cooling are targeted toward the successful use of sprinklers and mist systems. One of the important processes is the interaction between the fuel surface and water droplets. Previous studies on droplet surface impingement that have been performed usually involve the impact of water droplets on a heated surface or a liquid film. Related studies were performed on wood room condition surfaces [38], [39], and heated surfaces [40]. Surprisingly, evaluation of droplet impingement on a burning wood surface is lacking. Examination of this process is needed to understand one of the main phases of fuel wetting by water-based systems.

1.4 Research scope and objectives

This current study is aimed at investigating the local behaviour of water droplets in fire extinguishment mechanisms. In particular, this study focuses on the qualitative and quantitative measurements of a water droplet interaction with smoke particles and solid fuel surfaces in a fire scenario. The main topics of this research are focused on fundamental small-scale processes by utilising various imaging techniques to study the static and dynamic behaviour of water droplet evaporation.

In order to achieve the aim of this research, the scope of specific objectives has been defined to enable careful experimental work and the following analytical investigation. The objectives presented in this thesis are:

- 1. Review the literature on fire extinguishment mechanisms when water is used as the main suppression media, practical systems, experimental investigations and evaporation processes.
- 2. Assess the viability of the available visualisation techniques for water interaction with smoke and fuel beds.
- 3. Develop experimental apparatus and methods suitable for the investigation of water droplet interaction with burning solids and flames.

- 4. Develop a water droplet generator with the ability to generate different droplet sizes and speeds.
- 5. Study water droplet evaporation and surface characteristics using different imagining techniques.
- 6. Conduct an experiment to study the effect of droplet impact on burning surfaces.
- 7. Investigate the dynamics of droplets impact on fuel beds, splash behaviour, cooling of fuel surface, and effect on pyrolysis.
- 8. Identify the characteristics of solid fuel surface at different burning stages.
- 9. Examine the evolution of thermal characteristics of a water droplet impacting solid fuel samples.

1.5 Thesis outline

This thesis comprises eight chapters. Chapter one gives a brief introduction of the research background, the aim and the specific objectives of this current study, and a thesis outline to guide its readers.

Chapter two contains a literature review on fire dynamics and the mechanisms of fire extinguishment. Water spray systems are discussed as water has been one of the most reliable and effective methods to control or suppress a fire. The evaporation of water and the effect of suspended particles is reviewed. Furthermore, experimental techniques on water evaporation, imaging, interfacial tension, droplet generation, and droplet impact investigation are explored.

In chapter three, various experimental techniques used in this study are detailed. Visual and thermal imaging are used in different setups are described. Image processing techniques used in droplet evaporation, surface evaluation, and impact studies are outlined. The calibration of thermal cameras in different infrared bands is also presented.

Chapter four investigates the effect of suspended particles on evaporating droplets. Droplet evaporation is studied under different conditions along with different particle suspension methods. Preliminary results are given on surface tension by evaluating droplet movement on the suspension fibre. Chapter five contains a detailed study of water surface tension using the pendant droplet method. Salt solutions are also investigated along with pure water droplets. The stability and detachment of water droplets are discussed.

Chapter six presents a detailed analysis of the phenomenological and morphological aspects of droplet impact on cold and burning wood samples using high-speed imaging. The chapter shows major droplet spread and evolution processes and the effect of droplet diameter and impact speed.

Chapter seven investigates surface temperature and characteristics during droplet impact, which supports the work presented in Chapter six. This chapter shows that surface features vary depending on the burning stage. Variation in surface roughness, spreading, and absorption patterns influenced the evaporation time substantially.

Chapter eight is the last and includes concluding remarks on the investigations presented in this thesis. In addition, future work recommendation is given to build and expand on the results of this current research.

Chapter 2

Literature Review

2.1 Introduction to fire dynamics

Understanding the nature of fire is essential in knowing better ways of protection against it. Fire is a remarkable chemical reaction example which originate from combustible solids, liquids and gases. The only other type of materials when looking through the window of fire is noncombustible materials. Materials such as wood, ethanol, and hydrogen are, obviously, combustible. On the other hand, granite, water, and argon are noncombustible.

Combustible materials vary in many aspects related to fire protection. These aspects include flame spread rapidity, oxidiser requirement per combustible mass, heat output, smoke formation, and ease of extinguishment [41]. Prior to the occurrence of a fire, fires can be reduced by using measures such as fire-retardant materials or change in materials structure [42] although not always possible, such as in the case of wildfires. Once a fire breaks, different materials are used to extinguish a fire, including water, foams, gases, and other special techniques [43].

Different mechanisms can be identified by looking at the behaviour of fire and materials used in fire extinguishment. These mechanisms are fuel removal, fuel cooling, oxidiser dilution, and interfering flame chemistry by chemicals that react with free radicals.

Water, the most regularly used material in fire extinguishment, is suitable for many types of fires. However, water can also react in unforeseen ways with specific combustibles. For instance, hydrogen is produced when water reacts with burning metals [44].

Various gases are generated by fire and some of which are toxic. Carbon monoxide is one example which is produced in dangerous concentrations. In addition, solid particles are also produced. The properties of such particles are essential for some devices, such as smoke detectors which depend on the particle size and smoke concentration [45]. The smoke properties are related to the combustible chemistry and the process of combustion.

Heat transfer and fluid mechanics in fires are involved in significant interrelation in addition to the chemistry of combustion. Investigating the detailed interaction of these processes and the influence on fire is the subject of fire dynamics.

2.1.1 Fire termination

Fire requires a high temperature so that combustion reactions continue to produce heat at a similar rate of dissipation. The heat transfer balance ensures that the reaction zone in a fire will maintain a high temperature and will not cool down. The general view is that this balance needs to be disturbed, such as removing flammables, to reduce the reaction zone temperature and extinguish the combustion.

Some of the heat in the reaction zone of a fire is usually being transferred to the surroundings. The environment is typically cooler; therefore, the rate of additional heat removal needed for extinguishment is less than the heat being generated [46].

Extinguishment can be achieved by reducing the temperature of the gaseous phase of the fire or the liquid or solid combustible fuel. In the latter situation, the production of flammable gases is ceased or at least reduced initially by cooling the fuel [47], [48]. In fact, this is considered the main mode of fire extinguishment when water is applied to a wood fire.

Extinguishment can also be achieved by introducing a shield between the fuel surface and the flame, for instance, by introducing a water-based foam layer. The foam layer not only absorbs some heat and reduces the fuel's temperature but also acts as a radiation shield which prevents the radiative transfer from the flames to the surface of the fuel [49], [50].

Preventing the supply of air to the flames also will lead to extinguishment. An alternative way is to dilute oxygen present in the air by introducing an external supply of nitrogen, carbon dioxide, water, or the products of the combustion itself [51], [52]. Some of the fuel can be separated from the fire in certain situations. For example, a piece of furniture can be removed from a compartment fire.

Flames are the gaseous phase where free radicals take part in the combustion chain reactions [53]. Some chemicals, when added to the flames, reduce the concentration of free radicals and lead to a decrease in the reactions. For example, chemicals containing bromine can react with hydroxide radicals, which are always present in a combustion chain reaction [54]. However, many chemicals have been phased out due to their adverse effect on the environment and the resulting toxic products [55].

2.1.2 Smoke

Smoke is a mixture of (1) particulates consisting of soot, semi-volatile organic compounds (SVOC), and solid inorganic compounds; and (2) non-particulates consisting of very volatile organic compounds, volatile organic compounds, and liquid and gaseous inorganic compounds [56]. The particles can be categorised into solid carbon particles known as soot, and liquid mist, which are formed as a result of the condensation of some gas molecules. Soot is usually characteristic of black smoke as shown in Figure 2.1, while the liquid mist is characteristic of coloured smoke. Some definitions of smoke also include the evolved gases as well.



Figure 2.1: Smoke during a large scale flashover fire extinguishing experiment [57]

Including gases in defining smoke is essential because condensation occurs in a later stage [58]. The liquid mist will continue to form larger droplets as the environment cools down. On the other hand, smoke emits strong thermal radiation and the mixture of soot particles, and liquid drops can play a significant role in defining the radiation.

The study of smoke is important due to the implication on fire spread, and toxicity [59], [60]. In addition, the visibility aspect when encountering smoke and its ability to block vision is also significant because it can affect rescue operations and escape times [61]. Nevertheless, smoke is also used as an indicator for fires in the working principle of fire detectors [45].

Clearly, the amount and nature of smoke generated in any fire vary not only because of the burning material but also the burning conditions [62]. For example, a fire in a compartment can exhibit an object burning to produce radiant heat that causes another object to increase in temperature and produce vapours. Also, materials can produce different amounts of smoke based on the ventilation conditions.

2.2 Wood combustion

In combustion science, wood is considered a solid fuel and comes in different types. The type of wood depends on its shape and composition, having undergone various physical and chemical processes, for instance, hardwood, charcoal, sheets, pellets, and wood dust, to name but a few. As a result, wood has been a widely abundant material used in many applications such as paper products, ropes, furniture, construction, and fuel.

Wood consists of organic substances and minerals. Carbon is the most abundant and usually makes up to 50% of wood mass. This is followed by oxygen, hydrogen, and nitrogen with mass percentages 43%, 6%, and 1%, respectively [63].

2.2.1 Wood thermal decomposition

A thorough discussion of wood thermal decomposition is complex due to its isotropic and inhomogeneous properties. The structure of fibres, which is seen as grain in the wood, indicates that thermal conductivity changes with direction. Pores found in the structure create an easy path for volatiles to flow more in one direction than in another. The nature of wood as an organic structure results in considerably different thermal behaviours between species, and even various samples from the same source can have differences in physical properties [46]. Therefore, investigations of the chemical and physical wood properties have a tendency to be scattered, a result not of inadequate studies but essentially a manifestation of the real nature of wood.

2.2.2 Constituents of wood

Wood is a combination of natural polymers with high molecular weight. The main components are mainly cellulose, hemicellulose, and lignin which have an approximate contribution of 50%, 25%, and 25%, respectively [64]. Overall, the decomposition of wood can be viewed as an accumulation of the distinctive behaviours of its components [65]. When wood is gradually heated, changes begin to appear in its structure. These changes are accelerated by increasing temperature, which indicates the decomposition of the main wood components to create volatile gases, tar, and char.

Table 2.1 lists the temperature range at which wood pyrolysis occurs. The literature states varying values for pyrolysis because the measurements depend on many factors, including wood type, heating rate, and component separation. The pyrolysis starts when hemicellulose decomposes at 150 °C, and completes at around 350 °C, approximately. At 275 °C, the cellulose starts to decompose and ends at a similar temperature as that of hemicellulose. Lignin decomposes at 250 °C, and continues to 500 °C.

Cellulose forms the majority of wood structure and is a long chain of the molecule $(C_6H_{10}O_5)$. Ultimately, 15% of the mass is retained as char at a temperature of 400 °C. Additional reactions can occur at higher temperatures resulting in a reduction of remaining char. Hemicellulose has a random amorphous structure and is abundant in shorter chains compared to cellulose. The decomposition proceeds until 350 °C, producing largely volatile gasses and a very small amount of char. Lignin is responsible for holding the strength of wood and is composed of many components. It yields a significant amount of char, and around 45% ends up as char at a temperature of 800 °C. Lignin contributes the most to the char produced as a layer during wood thermal decomposition [66].

Component	Temperature range (°C)
Hemicellulose	150 - 350
Cellulose	275 - 350
Lignin	250 - 500

Table 2.1: Pyrolysis of wood components [67], [68], [69], [70].

2.2.3 Surface topography and structural effect

A slab of wood in its natural state has a defined surface, and the heat transfer can be understood in the usual manner. Heat transfer in wood is affected by the anisotropy in thermal conductivity. Studies found that the thermal conductivity along the fibres can be as twice that perpendicular to the grains. Furthermore, grains allow considerable gas permeability as shown in Figure 2.2.



Figure 2.2: Illustration of pyrolyzing layer of wood [64]

The post pyrolysed state of the surface layer is more complicated. The heat transfer and heat conduction are affected by a network of cracks that initiates on the surface at a certain stage of the heating. These cracks continue to propagate along with the pyrolysis wave. Heat transfer does not pass through a plane surface in the fissured zone. A convective flow or radiation of hot gasses can penetrate through the cracks on the surface. The heat is transported more rapidly into the layers, although the total heat transfer may be unaffected [43].

In the case of the wood grain being parallel to the surface, a tendency arises for volatile to escape to the fissures. The volatiles travel approximately at isotherms and then into wide cracks, convective heat transfer between hot gases and the solid will be unimportant in this case [71]. Volatiles are restrained by structure in the unfissured zone. In this region, pressure increases with distance and then decreases to the level of unaffected wood. The pressure distribution will lead the majority of volatiles to flow to the surface and the rest to the interior. Good thermal conductivity between the moving gases and the wood fibres results in a considerable convective heat transfer. This allows the assumption that the temperature distribution in the solid is the same as that in the gas.

2.2.4 Extinguishment mechanisms

Rasbash et al. [8] in a series of publications during the mid-twentieth century is one example of initial investigations on fire extinguishment. Early works on fire termination, such as that of Rasbash et al, identified the major extinguishment principles and were later classified into three main mechanisms [72]. The following mechanisms were recognised:

- Heat extraction, also sometimes referred to as cooling [6].
- Oxygen dilution, also referred to as oxygen displacement [4].
- Blocking of radiation, also known as radiation attenuation [73].

Complex mass and heat transfer govern all the mechanisms mentioned above when water droplets are applied to a fire environment. Other mechanisms were later considered, one of which is fuel bed covering [51]. The distinctive aspects of each mechanism do not mean that only one of them is applicable at once, but rather they are all involved to some degree in each situation of extinguishment. The measurement during fire proved to be difficult, and so the extent of each mechanism's participation has not been investigated comprehensively.

One of the cooling mechanisms is achieved by water droplets according to their size and momentum. Droplets with high momentum and size can usually penetrate the plume and reach the fuel bed to act on fuel surface and surrounding materials exposed to the fire.

The evaporation of small droplet in the gas phase cool the hot smoke and flame zone. In a compartment fire, cooling the gas phase reduces the use of unnecessary sprinklers [74], [75], [76].

Small droplets of water mists create a substantial liquid interface for heat absorption. Hence, gas phase cooling in water mists is considered an essential mechanism. In addition, water vapour expands and the effect of oxygen dilution is observed [10], [6], [72]. Theoretical estimation of evaporation cooling effect was found to be negligible for droplet sizes greater than 0.5 mm [77].

Figure 2.3 presents an illustration of oxygen displacement and dilution by water vapour. Experiments were conducted to study gas cooling, and displacement of oxygen with 250 µm mist in an enclosure fire [78]. The flame was a diffusion methane flame; both mechanisms were shown to be insignificant. But the results were attributed to the path of the droplet out of the plume rather than insufficient



Figure 2.3: Illustration of oxygen by water evaporation

evaporation. Other experiments confirmed that oxygen dilution is not significant in compartment fires [78], [79], [80], [74], [81]. On the contrary, experiments showed that a higher probability of extinguishment was accomplished when droplets were introduced to the combustion zone or the hot smoke layer [4], [82], [47], [83].

Radiation attenuation by water droplets is their ability to absorb radiative heat and reduce the thermal feedback to fuel beds. Figure 2.4 illustrates the water's ability to absorb and attenuate radiation. This effect was shown to be insignificant in reducing the radiative heat transfer to burning surfaces, and adjacent unburned surfaces [72]. A difficulty in assessing the contribution of radiative attenuation is that it cannot be separated from the overall cooling [78]. Theoretical analysis resulted in the estimation of water droplet absorptivity [84], [85], [86], [87]. The attenuation of radiative heat was compared to droplet size and overall spray loading. The absorption was significantly less than the scattering occurring on the surface of droplets. For a substantial blocking to take effect, droplet size needs to be particularly small with diameters less than 50 µm [72] since the attenuation factor correlated with the incident wavelength in the same order of droplet size. Slightly larger droplets in the order 100 µm can still play a role in evaporation by radiation absorption given an increased application flow rate [84].

Direct impact of water on fuel bed act as a cover to cool and wet the surface. Experiments of small scale liquid fuel interaction with droplets were qualitatively discussed [83]. In addition, solid fuel beds were also examined and the relation between



Figure 2.4: Illustration of radiation attenuation by water droplets

droplet size and flow rate were correlated with extinguishment time [88]. The main conclusion was that shorter suppression times were achieved with smaller droplets. Overall, larger droplets have a better ability in terms of penetration in large scale fires that are being controlled by sprinkler systems [89], [76], [90]. Usually this is referred to the issue of fire scale which has not been investigated in many fire cases [91].

In conclusion, the pioneering work of Rasbash et al. [8] and other early investigations on fire extinguishment laid the foundation for our understanding of the major extinguishment mechanisms, including heat extraction, oxygen dilution, and radiation blocking. However, the complexity of mass and heat transfer processes involved in these mechanisms, especially when water droplets are applied to a fire environment, has made it difficult to comprehensively investigate the extent of each mechanism's participation. Moreover, the interplay between droplet size, momentum, and fire scale remains an area of ongoing research, with numerous studies exploring the roles of small and large droplets in various fire suppression scenarios.

Despite the challenges in assessing the contribution of individual extinguishment mechanisms, progress has been made in understanding the importance of droplet size, momentum, and flow rate in fire suppression. Smaller droplets have been shown to be effective in gas phase cooling and radiation attenuation, while larger droplets



Figure 2.5: Spectrum of drop diameters that occurs in industrial systems and natural phenomena. Adapted from [96].

can penetrate plumes and reach the fuel bed, providing surface cooling and wetting. Ultimately, the development of more effective firefighting strategies will continue to rely on a deeper understanding of these complex interactions, as well as the behaviour of water droplets in diverse fire scenarios.

2.3 Water spray systems

The first pilot water spray in record was fitted in the Theatre Royal, London [92]. The potential of the system to save lives and protect properties was immediately recognised. Currently, systems of water sprays and sprinklers are widely used as the most effective and efficient method of fire extinguishment.

Since the nineteenth century, different bodies and organisations have played a role in how spray systems are regulated. The development of standards and rules for water spray system installations was initially driven by insurance companies such as Factory Mutual, the predecessor of FM Global [93]. This was soon carried out by research institutes and organisations to study many aspects of fire phenomena, from investigating different fuel types, and understanding the interaction of chemical and physical processes involved when the spray is applied, to exploring new additives to enhance water sprays. Organisations such as Fire Research Station was established in 1947 as a part of the Building Research Establishment in the UK [94], and Fire Research Centre in 1974 at the National Bureau of Standard (NBS), which later became known as the National Institute of Standards and Technology (NIST) in the USA [95].

A spray can be defined as a system of moving liquid droplets in a continuous fluid medium. Common examples of sprays are waterfall mist and rain. Other types of sprays that can be created by atomisers on purpose are paint spray and fountain spray. Figure 2.5 shows the drop size range that occurs in nature and others created by industrial atomisers. Examples of sprays that happen naturally are drizzle and rain.
Spray characteristics influence the behaviour of water in suppressing fire. The characteristics are dependent on the production technique, and therefore different methods will produce water sprays with different characteristics.

The atomisation of water droplets in a spray system leads to very small droplet sizes. Small-sized drops have a high surface area-to-volume ratio. An increase in this ratio enhances the ability of water to extract heat in the smoke layer, hot regions, and flame. Table 2.2 shows the increase of surface to area to volume with finer droplets.

Droplet diameter (mm)	Surface area of a single droplet (mm^2)	Volume of a single droplet (mm ³)	Surface area to volume ratio (1/mm)	Total surface area for 1 m^3 of spray (m^2)
1	3.14	0.52360	6	6000
0.5	0.79	0.06545	12	12000
0.2	0.13	0.00418	30	30000
0.1	0.03	0.00052	60	60000

Table 2.2: Surface area and volume of fine droplets

Higher surface area available for evaporation can reduce the smoke and flame zone temperature. Once the temperature of the flame is lowered below the limiting adiabatic flame temperature, which is essential to sustain the combustion, the flame will be suppressed. For most organic vapours and hydrocarbons, the critical temperature is 1326 °C (1600 K), approximately [43]. The reduction in flame temperature also decreases the radiative heat transfer from the smoke and flame zone to the fuel bed, hence decreasing the pyrolysis rate of the fuel, and this feedback reaches the extinguishment limit.

Several factors have been identified that affect the evaporation rate of water droplets. The factors are environment temperature, droplet surface area, heat transfer coefficient and the relative velocity of droplets in the surrounding gas [97]. The effect of micro-scale droplets on the methane-air burning velocity was investigated by Chelliah [9]. Chelliah compared the suppression effect of water with other gaseous and chemical agents. The diameter of the water mist was 1 µm and less. The conclusion was that water performed better than N₂ and CF₄ by more than three times in terms of used mass and performed twice as efficient at reducing the burning velocity.

Spray characteristics, influenced by the production technique, play a crucial role in the behaviour of water in suppressing fires. Smaller droplets, with their high surface area-to-volume ratio, are particularly effective at extracting heat from smoke layers, hot regions, and flames. Evaporation of these droplets can significantly reduce smoke and flame zone temperatures, eventually leading to flame suppression.

Research on the factors that affect the evaporation rate of water droplets and their impact on fire suppression has shown that water can be more effective than other gaseous and chemical agents when applied in the form of fine mists. However, further investigation is needed to continue improving our understanding of water spray systems and their characteristics in order to optimise fire suppression techniques and ensure the safety of lives and property.

2.4 Droplet evaporation processes

Evaporation of liquid droplets is a phenomenon that occurs naturally when the surrounding environment in the near vicinity of the droplet is not fully saturated with the same medium of the droplet [98]. In fact, droplets in the micro and nanoscale experience an increase in vapour pressure at the surface, which can lead to evaporation even in saturated environments. This effect of vapour pressure increase is due to the curvature of the droplet surface, which is referred to Kelvin's effect [99].

The examination of spherical droplet evaporation controlled by diffusion in a still environment is a less complex system that drop evaporation under convective flow. Moreover, the understanding of spherical droplet evaporation can be compared to non-spherical droplet behaviour so that the shape influence can be evaluated. Additionally, multicomponent droplets formed by mixing different foaming agents or suppression enhancers and a base liquid are of importance in firefighting. As a result, studying a droplet of a single component will result in a better approach to exploring multicomponent droplets. However, multicomponent droplets usually experience preferential evaporation of the component with the higher volatility [100], which makes these droplets more complicated.

Maxwell considered the evaporation of a droplet a process controlled by diffusion [101]. Maxwell considered the case where a droplet is placed in a motionless infinite environment. The first interesting evidence was shown by Morse [102] when he found experimentally that the evaporation rate was proportional to the radius of a drop and not the surface area. Using an analogy between heat conduction and diffusion, Langmuir [103] analysed the data of Morse and confirmed the results. Langmuir also concluded that the exchange of molecules from the drop to the interface in contact with air is rapid compared to the diffusion of vapour to the surroundings. Early work of droplet evaporation that was conducted by Morse, Langmuir, and others involved iodine droplets. Iodine has a relatively low vapour pressure and slow evaporation. Meanwhile, other researchers started to study the evaporation of micro-scale droplets and investigated the vapour layer effect at the surface [39], [104], [99].

Fuchs postulated that diffusion occurs at a distance of the mean free path away from the surface rather than at the surface immediately [105]. This distance means an introduction of a surrounding vacuous sphere around the drop. The idea of this sphere is particularly important for low-pressure and micro-scale droplets because the number of molecules available in this shell is relatively low. Several research studies then emerged to confirm the results of Fuchs [106]. Experiments on small water droplets suspended on thermocouple beads showed that Fucks accurately described the relationship between the vapour density difference of the surface and surroundings with the temperature difference [107]. New experimental results later explored the effect of surface cooling in more volatile drops than water, especially for large drops in still air [108] and in free fall water drops [109]. In fact, Fuchs revisited his initial publications and later analysed major results in evaporation, including derivation of Maxwell Stefan drop equations, the effect of absorbing vessels used in experiments, and fall in temperature of suspended and falling drops.

Evaporation of falling drops can be enhanced by convection. Convection is a boundary phenomenon where heat transfer occurs between the boundary and a moving fluid. The heat transfer at the boundary is determined by the thermal conductivity of the moving fluid and the temperature gradient in the vicinity of the moving fluid layer on the boundary.

Fine water droplets suspended in the gas phase of a fire can attenuate thermal radiation. Droplets act as a thermal barrier and reduce the thermal feedback to the fuel source and surroundings. Additionally, the water vapour in the smoke and flame zone, resulting from droplet evaporation, absorbs radiant heat from the flames and hot smoke. The first experiments on water mist showed a 70% reduction of radiative heat flux to the walls in a compartment test fire after mist activation [110]. The ability of water droplets to attenuate the thermal radiation in fires has been documented and recognised for shielding objects, properties, and people [111], but was not accounted for the effect in fire extinguishment [112], [48]. A system of water spray used in fire is considered a two-phase system with semi-transparent properties. It is a collection of liquid droplets, water vapour, and other gases such as carbon dioxide. Recent investigations used thermal imaging to study infrared attenuation of water sprays [113]. The need for droplet penetration into a plume means that

millimetre droplet size is needed and is much better suited for cooling the bed [48]. On the other hand, micro-scale fine droplets are more efficient in radiation attenuation [73]. Therefore, understating droplet evaporative properties in different situations is undoubtedly of importance when it comes to accounting for the different aspects of fire extinguishment mechanisms.

Water is semi-transparent to radiation as opposed to opaque liquids [87], [86]. Opaque materials absorb radiation on the surface; however, water experience a volumetric absorption. This is the most complicated aspect of radiation analysis for a water droplet in a hot environment [85].

Tesng et al. [85] considered the evaporation of a single droplet with convective and radiative heating, and the internal circulation was neglected by accounted by the concept of effective thermal conductivity. The study concluded that radiation increases the surface and internal temperature of the droplet when the relative importance between radiation and convection is significant. This was typical for droplets larger than 0.1 mm and higher ambient temperatures.

Evaporation of water droplets in high-temperature environments has indicated the emergence of a vapour layer. This layer was correlated with significant changes in the heat transfer between the droplet and hot surroundings [114], [115] .In addition, droplet temperature distribution has been shown to be inhomogeneous for an extended period of evaporation time [114]. The temperature fields across different planes in a droplet that is intensely evaporated indicated results of different evaporation rates based on the dominant heating mechanism [115]. When any of the conductive, convective, and radiative heat mechanisms are neglected in rapidly heated droplets, temperature variations in different sections of a droplet can reach 30-50% away from numerical results [116].

The reports on radiative heat flux are usually measured in terms of total radiative flux or directional radiation intensity. Wide angle sensors and scanning radiometers are used to study the emission from hydrocarbon flames [117], [118]. Other techniques such as Fourier transform infrared spectroscopy have also been used, as reviewed by Sacadura et al. [111], and Parent et al. [119]. Table 2.3 lists experiments of droplet evaporation at different conditions. Suspension methods are further discussed later in this chapter.

In summary, the process of liquid droplet evaporation is intricate and influenced by a multitude of factors, such as the size of the droplets, the surrounding environmental conditions, and the droplet composition. The capacity of water droplets to reduce thermal radiation in fires is well established and plays a vital role in extinguishing

Environment	Suspension material	Material size (mm)	Droplet size (mm)	References
Enclosed still air, controlled temperature	Glass fibre, thermocouple	0.1, 0.08	1.5	[108]
Steam flow with controlled humidity	Horizontal glass fibre	0.03-0.07	0.2-1.1	[15]
Preheated air water stream with controlled humidity	Vertical thermocouple bead	0.05	0.95-1.18	[11]
Hot air water stream. Controlled humidity	Vertical quartz	0.2	0.9	[120], [121]
Hot air stream, humidity not controlled	Vertical glass	0.1	1	[122]
Hot air stream, humidity not controlled	Hollow steel rod	0.8	2.67-3.37	$[16], [123], \\ [12], [124], \\ [125]$
Controlled temperature and humidity	Polypropylene thread	0.15	1	[126], [127]
Dry air flow	Asbestos thread	0.25	2	[128]

Table 2.3: Suspension methods in droplet evaporation experiments

fires. Nevertheless, it is essential to comprehend droplet evaporation properties under various circumstances to optimize fire suppression techniques.

This highlights the need for further research into individual water droplets, the impact of surface smoke particles on the droplet surface, and the interaction between the droplet and the burning surfaces of solid fuels like wood. The insights gained from these studies will be invaluable in enhancing the efficacy of water spray systems and ensuring the safety of lives and property.

2.5 Effects on droplet evaporation

2.5.1 Droplet size

Droplet size varies in water spray systems, as was shown in section 2.3. Experimentally, the design and operational parameters of a droplet generator will also result in a change of a produced droplet size [129]. When the assumption of a constant evaporation rate is made, the lifetime is then proportional to the square of the initial droplet size. This assumption also implies a linear change in the receding surface area of the droplet. Moreover, inner medium flow and temperature gradients can be significant for large droplets. The Biot number is a ratio of the heat convection on a boundary to the heat conduction in a medium, and it is usually used as a measure of the significance of temperature gradients.

$$Bi = \frac{hL}{k} \tag{2.1}$$

where h (W/m²K) is the heat transfer coefficient, k (W/mK) is the thermal conductivity of the droplet liquid, and L (m) is a characteristic length, in this case, L is the diameter of the droplet. For a low Biot number $Bi \ll 1$, the temperature gradient across a droplet is negligible and the temperature can be assumed uniform.

2.5.2 Droplet shape

Droplet shape stability depends on the dynamic forces imposed on the droplet. The inertial forces can lead to shape deformation so that a droplet flattens. On the other hand, the droplet interfacial tension causes the droplet to be spherical due to the tendency of surface energy minimisation. The Bond number quantifies the balance between the inertial forces and interfacial tension.

$$Bo = \left(\frac{r_d}{\lambda_c}\right)^2 \tag{2.2}$$

where r_d (m) is the droplet radius and λ_c is the capillary length given by:

$$\lambda_c = \sqrt{\frac{\gamma}{\Delta \rho g}} \tag{2.3}$$

where γ (N/m) is the interfacial tension, $\Delta \rho$ (kg/m³) is the density difference of the droplet and the surrounding medium, and g (m/s²) is the acceleration due to gravity [130]. For higher values of density and lower interfacial tension, the droplet shape becomes more prone to inertial forces. Therefore, larger droplets experience shape deformation more evidently.

2.5.3 Temperature and humidity

Environment temperature is one of the most significant factors in the evaporation process. The evaporation rate is governed by the heat transfer from the gaseous surroundings to the droplet interface. The heat transfer to the droplet is proportional to the difference in temperature between the ambient gas and droplet temperature. The temperature not only affects the evaporation rate but also the motion of any suspended molecules. Furthermore, an increase in the temperature difference leads to a higher Pe number. This increase results in a faster accumulation of suspended particles near the surface.

The humidity during droplet evaporation has an effect on the temperature distribution in a droplet. The relative humidity of the air-water mixture is the ratio of water vapour partial pressure to the equilibrium water vapour pressure. In other words, relative humidity is the amount of water vapour in the air to the saturated state of the air-water vapour mixture.

Gregson et al. [131] explored the influence of varying relative humidity on the evaporation of NaCl solution droplets. They found that NaCl starts to effloresce at a relative humidity of 45%. Increasing relative humidity beyond this value leads to the deliquescence of the solid particles. This shows that relative humidity can affect the physical phenomena occurring in the droplet.

2.5.4 Presence of particles

A major part of the literature on the effect of particles on surface and morphology is dedicated towards sessile droplets on a heated surface [98], [132], [133], [134]. The evaporation of sessile droplets is controlled by pinning effects, for instance, once a droplet is deposited on a solid surface, the contact line limits the wetted area. During the sessile droplet evolution, the contact angle varies with time depending on surrounding conditions and vapour pressure. Chon et al. [134]. indicated that droplet evolution during the evaporation did not experience a constant evaporation rate and explained that the reason is the changes in latent heat of vaporisation.

Wei et al. [135] investigated the effect of particle aggregation and accumulation on the interface of water droplets. In the study, the Peclet number Pe, which is the ratio of particle diffusion to droplet lifetime, was examined with initial particle concentration. In conclusion, the tendency of shell formation as a droplet evaporates was correlated with a reduction in evaporation rate. Peclet number is defined as follows:

$$Pe = \frac{d_d J}{D_i} \tag{2.4}$$

where d_d (m) is the droplet diameter, J (m/s) is the evaporative flux or the rate of volume loss per unit area, and D_i (m²/s) is the diffusion coefficient of particles in the base liquid. One model of the diffusion coefficient is Stokes-Einstein equation which shows the factors that affect the mobility of the particles:

$$D_i = \frac{k_b T}{6\pi\eta r} \tag{2.5}$$

where k_b (J/K) is Boltzmann constant, T (K) is the temperature of the fluid, η (Pa · s or kg/(m · s)) is the viscosity of the fluid, and r (m) is the hydrodynamic radius of a particle in the fluid. The *Pe* number indicates whether the evaporation is faster than the diffusion of particles and vice versa. For a given *Pe* number, information about the radial distribution of particles can be predicted during a droplet evaporation.

Similar to the temperature gradients, molecules of different materials in a liquid droplet can experience a driving force towards regions of low concentrations. Given enough time, molecules will diffuse in a given liquid droplet so that the concentration becomes homogeneous. This is particularly true for materials that can be dissolved [136]. A similar trend has been observed with insoluble particles as shown in Figure 2.6. For large Pe values $Pe \gg 1$, the evaporation rate is faster than the diffusion of particles into the droplet. As a result, the concentration at the surface increases rapidly. Even though a concentration gradient is generated, the effect of this gradient is minimal. Ultimately, a skin made of particles on the surface is formed which indicates a sufficient quantity of particles for the crust. The evaporation rate is expected to decrease in the latter stage because of the resistance introduced by the forming skin [135].

On the other hand, for low Pe values $Pe \ll 1$, the diffusion of the particles is adequate to prevent the accumulation of the particles at the receding surface. Hence, the concentration and dispersion of the particles continue to be homogeneous in the droplet. Figure 2.6 is a diagram representing the two cases discussed above. Droplets might experience changing Pe numbers during their evolution. Nevertheless, this aspect of particle accumulation and diffusion enables physical interpretation of the effects on the evaporation of droplets.

Water droplets and smoke particles act as scattering media during fire suppression. Water sprayers serve as a method to effectively contain thermal radiation in fires. Water, as the intermediate zone between a fire and surrounding objects, can shield the radiation and reduce the potential of fire spread. The ability of water to shield radiation under intense fire radiation has been studied to evaluate the use of water curtains [84], [138], [139], [140]. Water spray can be applied using many pieces



Figure 2.6: Illustration of single droplet evaporation with suspended particles. Adapted from [137], [135], [136].

of equipment in firefighting, such as sprinklers, water curtains in industrial facilities, drencher systems, offshore facilities, and ships [3]. Soot formed in fires acts as a scattering material in a similar manner to water droplets. Considerable studies have been conducted to understand the morphological and optical properties of soot [111]. For example, refractive indices of soot have been experimentally measured in the visible wavelengths [141]. The distribution of particle sizes under different combustion conditions has been the subject of many studies. The concentration and size distribution of liquid and gaseous fuels have been investigated [142], [143], [144], [145], [146]. The soot particles had sizes of 10-100 nm.

In Gan et al. studies of radiation effect on the evaporation of droplets suspended with nanoparticles, the particles enhanced the evaporation under visible radiation [147], [148]. The additional absorption of radiation by the particles was deemed to be the reason for faster evaporation rates.

Tanvir et al. [149] used IR radiation to study the effect of particles on the evaporation of ethanol droplets. They concluded that the deviation from the d-squared law increases with particle concentration. Two competing mechanisms were suggested that control the evaporation rate, namely temperature rise at the surface and particle accumulation at the surface. During the initial stage of evaporation, the first mechanism plays a role in absorbing radiation and enhancing the rate. However, later stages experienced a high particle concentration on the droplet surface and reduced the available effective area for evaporation. A similar study on evaporation by heated flow was packed by a similar argument [100].

Tuning the properties of the particles can give the ability to change the scattering and absorption behaviour. Properties such as size, shape, and concentration cannot be controlled in a fire scenario similar to that of specifically manufactured particles. However, there are certain shared aspects of these systems that can be comparable. For example, if the concentration of particles is very high, the incoming radiation will be absorbed in the thin surface layer. While at the other end of low concentration, not all the radiation will be absorbed [150].

Several works on droplets with diluted particles reported the existence of internal flows in droplets [135], and the presence of thermal waves [151]. In addition, particles dispersed in liquids lead to a change in thermophysical properties such as viscosity, heat capacity, and thermal conductivity [152].

Rasid and Zhang [27] examined surface contamination in fuel and reported the influence of nanoparticles on thermo-physical properties, finding that surface soot contamination significantly altered diesel droplet burning characteristics. They showed that evaporation and burning rates were reduced and stability critically affected. However, they focused on burning droplet behaviour without further experiments on evaporation as an individual process.

Certainly, the problem is complex due to the variables of smoke content, particle size distribution, particle agglomeration, chemicals reaction with water, and the type of forces that arise between water and contamination. Considerable research has been conducted on smoke particles, the interaction between fire and water spray, and nanoparticle effect on fluids. However, so far, no experimental investigation has been conducted into the fundamental properties of smoke particle contaminated water droplets. Therefore, one objective is to investigate the influence on evaporation rate and surface tension of water droplets. Water spray has a significantly larger surface area than that of a single droplet having a comparable mass. Therefore, the possibility of contamination is much higher in real applications compared to a single droplet test. However, studying a single droplet system enables enhanced magnified imaging throughout the droplet lifetime.

2.6 Experimental techniques

2.6.1 Interfacial tension

Interfacial tension is relevant to many processes [153], [154], [50] when the capillary length become the dominant scale. The origin of interfacial tensions is the unbalanced intermolecular forces near the interface of two meeting fluids [155]A molecule in a liquid experiences cohesive forces by neighbouring molecules. These attractions result in a zero net force apart from those on the surfaces where another type of molecule is abundant. The forces acting on different types of molecules over the sides of a surface are called adhesive forces. This phenomenon is manifested by a tendency to have minimum surface energy leading to a favourable surface area. Interfacial tension not only governs the shape of drops, but also influences the wetting [156], drop surface impact [39], coalescence [88], and spray formation. Accurate measurement of the interfacial tension is therefore of fundamental significance in understanding systems such as emulsions and suspension drops.

Many methods have been used to measure interfacial tension as shown in Figure 2.7. The principle of the method has been discussed in the literature [157], [155], [158].Wilhelmy plate and Du Nouya ring are direct techniques based on a microbalance for interfacial tension measurement. The fine liquid layer which is produced when either the plate or the ring is pulled further away from the liquid bulk, can be related to the force of the microbalance. The bubble pressure and the growing drop/bubble methods utilise the internal pressure, and the radius of the bubble to find the interfacial tension. Capillary rise and drop volume methods are based on the balance between gravity and capillary forces.

Different interfacial tension measurement techniques are reviewed in Table 2.4. The table includes accuracy, time range for dynamic systems, surfactant suitability, two liquid suitability, viscous suitability, the possibility of particle application on the interface, and further notes. The accuracy of these methods for gas-liquid interfaces is 0.1 mN m^{-1} . The capillary rise method can achieve considerably better accuracy than other techniques in Table 2.4.

Most of the interfacial measurement methods in Table 2.4 have been used successfully for two liquid interfaces. Both Wilhelmy plate and Du Nouy ring method experience difficulties in evaluating the weight of the attachment in the lower density liquid. All methods result in lower accuracy when utilised for two liquid systems as well as when applied for viscous fluids.

Many difficulties face the measurement of viscous liquid interface tensions. Not only the slow flow velocity and long viscous deformation time are among the problems, but also some practical considerations such as liquid handling, injection, and application of the required volume. Methods such as the sessile droplet technique are recommended for viscous liquids because a droplet can be allowed for hours to reach equilibrium before taking the measurements. Sessile and pendant drop techniques are based on the balance of gravity and surface tension forces, but additional analysis of the shape distortion is required for measurements. Table 2.5 summarises different fitting approaches to extract surface tension from droplet shape when using pendant droplet method. The spinning drop and micropipette methods are based on distortion by evaluating the centrifugal force in the first method and the pressure required to distort the droplet in the latter.

Surfactants act as a part of the interface when they are introduced to a liquid and they will affect the interfacial tension measurements. Therefore, attachments such as needles, rings, and capillary tubes must be cleaned before attempting any measurements. In addition, impurities can adsorb on the liquid interface and solid attachments and therefore affect the measured results.

In dynamic systems with surface active solutes, interfaces that have been newly created are generally not in equilibrium. The interface will reach an equilibrium state after solute adsorption and distribution. The measurements of such surface behaviour is known as dynamic surface tension. Dukhin et al. [157] and Joos [159] described the theoretical background of dynamic surface tension of surfactant solutions. Table 2.4 lists characteristic time range for the mentioned techniques [160], [157], [161], [158]. Interfacial tension can vary over at least several seconds, and this change can continue for several hours.



Figure 2.7: Illustration of various interfacial tension measurement techniques.

Method	Accuracy	Time range	Surfactant Suitability	Two liquids suitability	Viscous liquid suitability	Particles application	Notes
Pendant drop	0.1	$> 10 \ { m s}$	Very good	Very Good	Very good	Possible	Surface active chemicals might release drop
Sessile drop	0.1	$> 10 \mathrm{~s}$	Good	Very Good	Good	Possible	Surface roughness affects the measurements
Bubble pressure	0.1 - 0.3	$1~\mathrm{ms}$ - $100~\mathrm{s}$	Very good	Very good	Good	Possible for volume mixtures	Hydrodynamic effects near the interface
Droplet volume	0.1-0.2	1 s - 20 m	Limited	Good	Inadvisable	Possible	Sensitive to vibration
Capillary rise	$\ll 0.1$	$> 10 \ { m s}$	Very good	Very good, experimen- tally difficult	Inadvisable	Difficult	Surfactants might change surface wetting properties
Wilhelmy plate	0.1	$> 10 \ s$	Limited	Good	Inadvisable	Difficult	Surfactants might change plate wetting properties
Du Nouy ring	0.1	$> 30 \ s$	Limited	Reduced accuracy	Inadvisable	Difficult	Same as above

Table 2, 1, 1 chormance of amercine surface tension teeningate	Table 2.4 :	Performance of	different	surface	tension	techniques
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Technique	Features	Notes	Reference
Arithmetic tabulation of geometrical function	Simple, Only two sections of a droplet are used	Tables were devised experimentally. Tables were later improved by numerical integration of Young Laplace equations	[162], [163]
A comparison between an actual droplet profile and theoretical profiles. The sum of squared residuals is found, the minimum sum represented the best fit.	Fourth order Runge Kutta integration of an approximate objective function	Objective function is simplified for ease of calculations. Drop width and height are required for initial guess	[164]
Optimisation scheme to reduce the sum of squared residuals	Uses Newton Raphson method along with incremental loading to solve the problem of initial guess	Does not require the apex, drop width, or drop height which leads to an increase in accuracy. The objective function was strictly the normal distance between measured and calculated profile.	[165]
Optimisation built on Rotenberg et al. [165]	Use of rotational discrimination instead of Newton Raphson method	4-50 times less processing time. Error analysis is performed.	[166]
Axisymmetric drop shape analysis	Different optimisation methods such as Levenberg Marquardt, steepest gradient descent	Different methods but all based on shape. Robust convergence. Objective function parameters similar to Jennings and Pallas [166]. Including scale, rotation, and other parameters	[167]

Table 2.5: Different fitting approaches to extract surface tension from droplet shape when using the pendant droplet method.

2.6.2 Impact studies

Several studies of drop impact on solid surface evaporation revealed that temperature variation could be generated at the interface. The Marangoni effect is a convection phenomenon that occurs as a result of gradients in surface tension and can lead to interface instability. Furthermore, when a drop is put on a heated surface, the considerable temperature difference between the drop and surface can lead to thermal and convective streams caused by both buoyancy and Marangoni effects. Experiments were conducted to investigate such convection streams by the use of infrared thermography in hanging drops [168]. Temperature measurement of the surface below the drop was obtained by thermo-liquid crystals [169] and infrared thermography [170].

Wetting and fuel surface cooling are among the different fire extinguishment mechanisms that have been investigated in hydrocarbon fires [37]. Surface cooling experiments are focused on the effective use of mist systems and water sprinklers. The interaction between water droplets and the fuel bed surface plays a significant role in cooling the fuel. Numerous studies on droplets' surface impact have been conducted. The experimental investigations usually involve drying water droplets on a heated surface or a liquid film. Table 2.6 lists relevant studies on wood conducted for room temperature surfaces [38], [171], and slightly heated surfaces [40]. Figure 2.8 shows a droplet evolution on a wood surface at 120 °C. Surprisingly, evaluation of droplet impact on a burning wood surface is lacking, which is required to understand the first stage of droplet evolution in a fire extinguishment by water-based systems.

Whether suppression by water systems can terminate a fire depends on many aspects that are usually investigated, such as the compartment geometry, spray location, nozzle operating pressure, droplet size distribution, and fuel type [72]. However, the local water surface interaction is barely investigated. The impact dynamics can play a major role in the heat exchange and mass transfer on the solid fuel bed.

Sprinkler fire suppression usually contains water droplet sizes in the scale of 1 mm [172]. Droplets of this size are different in terms of size when compared to the optimal size for water mist-based systems, which are usually in micrometre scale [9]. Large droplets enter the fire plume with an insignificant contribution to the gas phase cooling. The primary reason is that droplet size and relatively brief residence time, which only leads to negligible evaporation. However, once the droplet impacts the fuel surface, the dominant mechanism of fire suppression is dependent on the interaction between the burning solid and liquid water [173].

Interaction of water droplets with a high-temperature gas zone environment or heated wall can occur under a variety of heating conditions. Primarily, the gas zone



Figure 2.8: A 2.4 mm droplet impact on heated wood surface at a velocity of 1.71 m/s and $W_e=97$ [40]. Here the impact Weber number can be defined as $W_e = (\rho V^2 D)/\sigma$, where D is the drop diameter, V is the drop impact velocity, ρ is the liquid density, and σ is the drop liquid surface tension. The colours represent the different phases of liquid and surrounding environment.

environment involves two heating mechanisms, namely convective and radiant heating. On the other hand, conductive heat transfer is significant for a droplet impacting on a hot surface when the liquid streams become negligible. Generally, a droplet undergoing an impingement on a solid surface might exhibit various situations or a combination of fuel surface deposition, absorption, splashing, initiating a layer of hot vapour at the interface, and rebounding [174], [31]. These scenarios lead to a change in the behaviour of water flow on the surface which is influenced not only by the physical properties of the drop but also by the condition of the target surface material.

Droplet evaporation on hot surfaces can experience different regimes. Measuring the evaporation of droplets of the same volume on a surface with varying temperatures is a common technique to determine evaporation regimes. Figure 2.9 shows four distinct regimes of heat transfer: film evaporation or single phase regime, nucleate boiling, transition boiling, and film boiling [175]. In the film evaporation regime, heat conduction through the liquid-solid interface is dissipated at the liquid gas interface by evaporation. In the nucleate boiling regime, bubbles are produced, and the heat transfer increases to a maximum which is the upper limit of this regime. In the transition boiling regime, intermittent vapour pockets are formed beneath the droplets leading to a heat flux decrease. The start of the film boiling regime is marked by the levitation of the droplet over a layer of vapour where the droplet's lifetime reaches a maximum. The starting temperature of the film boiling regime is referred to as Leidenfrost point [176].



Figure 2.9: Regimes of droplet evaporation on a hot surface. Adapted from [177], [175], [178].

Water impact on organic materials characterised by the roughness due to their grooves has been rarely reported in the literature. Wood with higher density was correlated with lower critical impact Weber number for droplet splashing [171]. Furthermore, Lan et al. [40] tested water impact on a heated wood surface and observed liquid phase deformation at the solid wall due to the heat transfer between the solid and the liquid drop. Conditions of splashing droplets over processed materials such as cardboard were also investigated [154]. Previously, the dynamic aspect of splashes on burning natural woods did not attract much research. However, the outcomes of this process might lead to important conclusions for the fire extinguishing process of wood constructions and objects.

The outcomes of these investigations are influenced by several aspects involving both surface and droplet characteristics. The factors include properties of the liquid such as density, surface tension, droplet diameter, impact velocity and direction. In addition, surface features such as wettability, roughness, and temperature play a key role in the process[31]. The phenomena associated with all these parameters require a group of these variables to be restricted when designing an experiment. A description of the behaviour of a water droplet impacting a burning surface is therefore needed.

Conditions	Droplet size (mm)	Impact velocity (m/s)	Reference
Room temperature	2.4	1.1-2.8	[171]
Heated wood 80-120 $^{\circ}\mathrm{C}$	2.4	1.7-2.8	[40]
Room temperature, water with additives	1.8-2.4	1.1-2.8	[179]
Room temperature, foam water solution	0.24	1.50-2.80	[156]

Table 2.6: Droplet impact on wood samples studies

2.6.3 Single droplet suspension methods

The evaporation of a single droplet can be influenced by the methods it is produced. The main techniques used to suspend single droplets are shown in Figure 2.10. Depending on the working principle of the technique, they can be categorised into three types, namely filament suspension, levitation, and fall techniques. In the case of filament suspension, the droplet is held by a fibre, glass, or even a thermocouple bead. In contrast, other suspension methods, such as levitation, use airflow as a dynamic balance or an acoustic levitator to make a droplet float [180]. Several researchers have reviewed the methods, explained the physical principles, and discussed their potential applications [136], [137].

Table 2.7 provides a summary of diverse methods, their working principle, merits and potential uses. The applicability of methods and their relevance to fire studies are discussed below [181], [182].



Figure 2.10: Illustration of single droplet evaporation techniques: fibre or filament suspension, acoustic levitation, free fall, and aerodynamic levitation.

Suspension of a droplet on the tip of a filament is an intrusive method. The filament can be of different types, namely glass filament, fibre, and thermocouple bead. The environment can be controlled, such as still air or air stream. Several parameters can be measured, including diameter, mass loss, and temperature. Droplet diameter can be evaluated by image processing of sequential images of droplet evolution. Mass loss can be inferred by the deflection of a suspension fibre as a cantilever or by using a precise mass balance. Alternatively, the diameter can be used to estimate the mass, including shape deformation. In fact, shape deformation due to the suspension object can impact droplet evaporation [183]. However, careful selection of the fibre material can reduce the effect. For example, a 0.8 mm droplet can experience 10% of the heat transfer originating from the suspension fibre [184].

Aerodynamic and acoustic levitation can be viewed as methods to reduce the intrusive natures of suspension techniques at first instance. Airflow can be used to balance the weight of a droplet. The principle of aerodynamic levitation is to produce a drag force capable of the droplet weight. However, this method has stability problem and usually face difficulties with upholding the droplet with motion [136].

Free fall or microgravity experiments are used to reduce the interference of a suspension filament, aerodynamic field, or acoustic field. As the name of the method indicates, the droplet is not suspended during evaporation but rather falls for a certain period depending on the available distance. Therefore, this method entails that droplet evaporation can be observed in similar conditions of water spray dynamics.

Method	Filament	Aerodynamic levitation	Acoustic levitation	Free fall
$\operatorname{Principle}$	Suspension on the tip of a fibre, glass filament, or thermocouple bead	Droplet weight is balanced by drag force of opposing air flow	Levitated by the pressure field of acoustic waves	Droplets descend- ing in a chamber
Measurements	Diameter, mass, temperature	Diameter, mass, temperature	Diameter, temperature	Indirect measurements
Merits	Measurements can be obtained simultaneously	Measurements can be obtained simultaneously	Measurements can be obtained simultaneously	Can be used with uniform hot flows
Issues	Intrusive, heat transfer through filament	Droplet position and shape can be unstable	Heat transfer and shape are affected considerably	Require many instruments to track droplet path
Applicability	Droplet size de- pends on filament diameter 0.5 – 9 mm	Flow conditions such as speed de- pend on droplet size	Vibration effect due to wave. Droplet size 0.3 - 1 mm	Air flow can be controlled when a chamber is used. Requires many cameras to record the dynamics with suitable resolution.

Table 2.7: Single droplet suspension methods: working principles, measurements, merits, issues, and applicability.

Figure 2.10 shows a schematic of this method that illustrates the main principle of free-falling droplets. An advantage of this method is that not only a single droplet can be observed, but also a stream of droplets with identical properties such as size can be monitored.

A disadvantage of microgravity experiments is that direct measurement of some parameters, such as temperature becomes impossible. However, other indirect techniques such as imaging can be used to monitor droplet evolution, and velocity [181].

2.7 Conclusions

In conclusion, the experimental methods used in the study of fire extinction and droplet evaporation are vital to understanding the mechanisms behind them. Fire requires a high temperature to maintain the reaction zone and must be disturbed to be extinguished. Extinguishment mechanisms include heat extraction, oxygen dilution, and blocking of radiation. Water spray systems are the most widely used and effective method of fire extinguishment. Droplet studies are fundamental to understanding the behaviour of water spray mechanisms in extinguishing fires.

Droplet evaporation is influenced by factors such as interfacial tension, the Marangoni effect, and temperature variations. The measurement of interfacial tension is fundamental in understanding systems such as emulsions and suspension drops. Techniques such as filament suspension, acoustics levitation, and free fall techniques are used to suspend single droplets and are essential in understanding the physical principles and potential applications. These experimental methods have provided critical insights into fire extinction and droplet evaporation and will continue to play a vital role in advancing research in these fields. In the following chapter, we present the experimental methods that have been developed and employed to investigate various aspects of droplet evaporation, droplet surface tension, and the interaction of droplets with burning wood surfaces.

Chapter 3

Experimental Methods and Materials

In this work, several setups have been developed and employed to investigate droplet evaporation, droplet surface tension, and the interaction of a droplet impacting a burning wood surface. Specifically, four different setups have been utilised and identified in Figure 3.1. The first setup was used to study the particle effect on droplet evaporation. The second setup was used to investigate the effect of particles on droplet surface tension. The third setup was employed to study the phenomenological aspects of droplet impact on burning wood surfaces. Finally, the fourth apparatus was used to characterise the thermal and surface characteristics of wood samples impacted by a single water droplet.



Figure 3.1: The experimental setups developed and utilised in investigating droplet interactions.

In these setups, the imaging techniques utilised can be grouped into visual and thermal techniques. The visual techniques include using cameras for tracking droplet evaporation and droplet spread upon impacting burning surfaces. Furthermore, visual techniques also include surface mapping based on focus variation. Thermal techniques involve using thermal cameras and sensors for thermal surface characterisation. These techniques are detailed in this chapter, while the following chapters explain the specification of each technique and the experimental results.

3.1 Droplet evaporation

In this work, water droplets were suspended using a 0.1 mm silicon carbide fibre with a 0.01 mm diameter tungsten core. According to the experiments conducted by Liu et al. [185], 0.014 mm fibre allows drop suspension of diameters less than 1 mm and 0.08 mm fibres enables the suspension of droplets with diameters around 1 mm. These experiments show that the ratio of suspended droplet diameter to fibre size is 10, approximately. The main focus of this current investigation is the droplet size evolution with time. Therefore, the selected fibre mentioned above is suitable for the suspension of a droplet with a size close to 1 mm. Figure 3.2 shows a suspended water droplet on a 0.1 mm silicon carbon fibre.



Figure 3.2: A suspended water droplet on a 0.1 mm silicon carbon fibre.

A microsyringe was used to inject a droplet on the intended location of the fibre. The syringe was used together with a hypodermic needle of 0.3 mm diameter and 13 mm length. The droplet was then viewed for real time measurement of its size to ensure repeatability and consistency.



Figure 3.3: Initial size variation of suspended droplets. The experiment was repeated 100 times to measure the standard deviation.

Figure 3.3 shows the initial diameter of 100 trials of suspended droplets. The deviation was kept as low as possible to ensure the repeatability of the experimental procedure. The average diameter of the water droplet was 1.081 mm while the standard deviation was 0.069. Thus, manually injecting a droplet can be justified by measuring the initial diameter at each trial.

3.1.1 Particle application

Two case studies were performed to show the effect of wood soot on water droplet evaporation. The first case was to pre-apply the soot on water droplets of 1 mm in diameter using the smoke of small wooden pieces. This was then compared to the evaporation of distilled water droplets without applying smoke particles.

In the second case, slightly larger wooden pieces were used to apply soot to droplets. Droplets were evaporated partially because of the heating by the effect of convection and radiation in this case because the wood pieces were larger. This was compared to droplets evaporated using a constant power heating source. The source was designed to keep its power constant during all tests. In each test, the process starts by supplying the same power to a stainless-steel heating element. The element was then allowed to reach a steady state. Then, the coil was moved under the droplet by a linear actuator. The actuator ensures the consistency of heating element placement under the droplet.

The first case data did not allow the determination of whether the smoke particles contributed significantly to droplet heating. Convective heating from smoke to particles depends on heat transfer coefficient and temperature difference. In the second study case, larger wood particles were used to contaminate water droplets. This case was conducted to investigate contamination process contribution to increased initial droplet temperature. Similar to the first case, the data from the second case were compared to pure water evaporation. However, heat element setting ensured pure droplets had higher surrounding temperature range than the smoke contaminated droplets. The methodology in the two cases mentioned above was later improved by designing a new setup. The improved apparatus is explained and detailed in Chapter 4.

The experiments with small and large wood samples and their soot particles are relevant to real fires because they provide insights into the behaviour of water droplets which are used in the form of sprays in firefighting. In a real fire, water droplets are sprayed onto a burning surface to extinguish or control the flames. However, the effectiveness of water droplets in extinguishing a fire depends on various factors, such as droplet size, surface tension, and the temperature and composition of the burning material.

The experimental methods described in the chapter aim to investigate the effect of wood soot particles on water droplet evaporation, as well as the role of droplet heating mechanisms in the presence of wood particles. The results of these experiments can help to better understand the behaviour of water droplets in real fire scenarios where wood soot is present and thus inform the development of more effective firefighting strategies. For example, the findings may suggest ways to modify the composition of water droplets or the methods of delivering them to the burning surface in order to optimise their effectiveness in extinguishing fires.

3.1.2 Image processing

Raw images require processing before segmentation can be applied. Images were analysed in Matlab with the aid of the Image Processing Toolbox. Figure 3.4 shows the steps in processing the images to extract the diameter.



Figure 3.4: Flow chart of image processing and data export.

The actual droplet size was calculated by finding the spatial resolution of the images. The spatial resolution can be found by calibrating the pixels that span across the fibre diameter to the diameter of the fibre.

Under normal gravity, suspended droplets take the shape of an ellipse. The shape segmented in Matlab is based on the major and minor diameters of the droplet. The equivalent volume is evaluated from the product of the major diameter times the square of the minor diameter. The equivalence of ellipsoidal and spherical volumes means that an equivalent droplet diameter can be calculated. Droplet diameter evaluation by using the assumption of the ellipsoidal and spherical volume equivalence was conducted by many researchers on droplet evaporation [132] [100].

3.1.3 Visual imaging

The imaging setup of droplet evaporation experiments in this present work is shown in Figure 3.5. The setup includes: a camera, micro lens, silicon fibre, heating element, light source, and a diffuser. The camera was Phantom SA4 and it was used to capture droplet evolution. For a magnified view, a Nikon AF Micro 60 mm f/2.8 lens was used along with an extension tube. A silicon carbide fibre was used to suspend a water droplet. The droplet was backlit by IDT LED illuminator. The light was diffused using a piece of white paper. Light diffusion allows the camera to capture clear images with low contrast between the droplet centre and circumference. Also, it prevents light source reflection on the droplet. A 1 ml syringe was used to inject a droplet and suspend it on the fibre. The fibre diameter was 100 µm. The initial droplet diameter was 1 ± 0.1 mm for all tests.



Figure 3.5: Visual imaging setup for water droplet evaporation.

3.2 Pendant drop tensiometry

Pendant droplet tensiometry is a method for measuring the interfacial tension of two mating fluids. For example, the fluids can be oil and water or air and water. The principle of the technique is based on the drop shape analysis, which is captured by imaging a backlit drop suspended by a vertical capillary. A single image can be used to extract drop surface area, volume, along with interfacial tension. Figure 3.6 shows a schematic of a droplet suspended by a capillary. The schematic indicates the droplet density ρ_d (kg m⁻³), ambient or gas density ρ_g (kg m⁻³), coordinates at the (r, z), radius of curvature at the apex R_0 , tangent angle ψ at any point s, and image misalignment angle ω .



Figure 3.6: A pendant drop schematic suspended by a capillary. The grey region indicates the captured area by a camera, which might be misaligned with the drop.

What makes the extraction of these values is the Young Laplace equation, which states the relation between the increase in pressure across the interface and the curvature at a point on the surface. The curvature is defined by the two principle radii, which are enough to describe a point at the surface. The Young Laplace equation is given by

$$\Delta P = \gamma \left(\frac{1}{R_{c,1}} + \frac{1}{R_{c,2}} \right) \tag{3.1}$$

Assuming that the pressure difference is due to the hydrostatic pressure, the droplet profile can be described by axisymmetric set of equations:

$$\frac{d\psi}{d\bar{s}} = 2 - \beta \bar{z} - \frac{\sin\psi}{\bar{r}} \tag{3.2}$$

$$\frac{d\bar{r}}{d\bar{s}} = \cos\psi \tag{3.3}$$

$$\frac{dz}{d\bar{s}} = \sin\psi \tag{3.4}$$

where z (m) is the vertical axis starting at the drop apex, r (m) is the horizontal axis, s (m) is the length of the arc along the profile starting at the apex, ψ (rad) is the tangent angle between the drop profile to the horizontal at any point along the interface, and β is the Bond number which contains the properties of the two fluids dictating the profile shape.

The Bond number is defined by

$$\beta = \frac{\Delta \rho g R_0^2}{\gamma} \tag{3.5}$$

where $\Delta \rho$ (kg/m³) is the difference in density between the fluids across the interface, g (m/s²) is the acceleration due to gravity, R_0 (m) is the radius at the apex of the drop.

The experimental setup needed to measure the surface tension of a pendant droplet is shown in Figure 3.7. The setup consisted of a camera, an LED light source with a diffuser, a peristaltic pump to control droplet volume, and blunt needles of different sizes. Special consideration must be given to ensure satisfactory quality of images for precise measurement of surface tension. Optical aberration and reflections can be avoided by appropriately diffusing the light and limiting surrounding light sources. Likewise, the analysis of images is considerably enhanced by ensuring a homogeneous background and using a lens with minimum distortion effects.



Figure 3.7: Schematic illustrating the pendant drop experimental setup.

A representative image of a pendant droplet suitable for measurements is shown in Figure 3.8. The vertical position of the needle is absolutely essential so that the droplet profile is axisymmetric. Different sizes of needles were tested to ensure that the shape of a droplet had negligible distortion because of gravity. If the shape of the droplet is only slightly distorted, it follows that the Bond number is low, and this can lead to inaccurate profile fitting.

Gravitational forces affect the shape of drop by elongation away from the spherical shape that tries to minimise the surface energy. An algorithm that solves the equations needs first to recover the profile of the interface. The interface can be extracted by any edge detection method and are usually implemented in many software packages. An example of a widely used edge detection algorithm is the well-documented and established Canny algorithm [186].

Following profile extraction from an image, the shape equations are solved and optimised for the best fit. The method requires the drop to be in equilibrium and shielded from any air fluctuation and rapid temperature variation. The light source should be carefully set for sharp identification of the interface, and that other reflected sources should be avoided. A good solution of the shape equations determines the Bond number, and then the surface area and volume can be calculated. The Bond number equation is then applied to find the interfacial tension, given that densities



Figure 3.8: Image of a pendant drop in pendant droplet tensiometry setup.

of fluids and gravity are known, while the radius at the apex is extracted from the image.

3.3 Droplet generation methods

Experiments involving single droplets require an efficient and reliable method of producing droplets. Droplet generation techniques vary significantly based on the application, droplet parameters and working fluid. The main distinction of interest can be made based on droplet size. The common way of producing millimetre scale droplets is by pressurised liquid flowing out of a small hole which will result in stream breaking into separate droplets. This is explained by the fact that inertial forces of the stream overcome the surface tension. On the other hand, micro scale droplet production usually requires overcoming the interfacial forces between the liquid and the ejection nozzle [30].

Another distinction that can be made is the two principal droplet generation modes. The droplets can either be produced in a continuous stream or individually. A stream of droplets is achieved by flowing fluid which is disturbed externally. The jet can be disturbed by a secondary air flow, vibrating part or acoustic pressure wave. In contrast, a droplet on demand is accomplished by a short duration flow. Piezoelectric elements is a common way implemented in droplet on demand devices. Table 3.1 is a list of droplet generators with a comparison of their performance.

Technique	Droplet Size (mm)	Nozzle size (mm) & type	Production rate	Mode	Reference
Piezoelectric element	0.65-1.32	1.02 3D printed ABS	Every 2 Seconds	On demand	[36]
Resistive heating of nozzle tip	0.11-2	0.11-0.31 Stainless Steel syringe tips	Injected by a syringe up to a specific hanging size	Single	[35]
Mechanical vibration	0.1-1	0.2-0 heat processed capillary glass	Every oscillation, Tested up to 5 kHz	Continuous stream	[34]
Solenoid impact	0.1-0.3	0.1-0.3 Sapphire	1Hz to tens of Hz (as stated)	On demand	[33]
Piezoelectric element	0.5-1.4	0.5-1.4 stainless steel	Not tested	On demand	[32]

Table 3.1: Comparison of various devices and their performance

Repeatable experiments require reliable droplet generation. Existing droplet on demand generators are highly priced, while most in house manufactured devices are complex and made for specific application. Several droplet generator novel designs have been proposed for a variety of reasons including construction simplification, enhanced control, disturbance reduction and high extreme working condition ability.

Kosch et. al. [34] used a hard drive actuator arm to vibrate a nozzle made from capillary glass. The assembly of the generator is shown in Figure 3.9 (b). The The nozzle produces droplets according to the vibrating frequency. However, the vibrating glass was liquid supplied by a syringe pump and only continuous droplets stream was produced rather than individual droplets. Although hard drives and capillary are inexpensive, both items required accurate modification.

Other designs implemented thin piezoelectric elements. Unique techniques were presented in order to reduce the cost such as using a 3D printed structure [36] and low voltage piezoelectric films for portable devices [187]. Nonetheless, Current designs of piezoelectric requires power amplifier and stable pulses and droplet size has a nonlinear relation with pulse width. In addition, piezoelectric designs require careful tuning and experimentation to produce same size droplets. Resistive heating was used to separate a droplet from an injection nozzle [35]. The droplet was monitored before separation, and the heating of the nozzle tip droplet interface was only applied once the required size droplet was hanging at the nozzle tip. While resistive heating was an innovative method, droplet shape was distorted occasionally, and secondary droplets were formed in other cases.



Figure 3.9: Schematic of droplet generators based on different techniques.

The techniques described in the passage are relevant in producing a novel droplet generator for combustion studies because they offer different methods of generating droplets with varying sizes and characteristics, which are necessary for studying the fire extinguishment of different materials. The use of thin piezoelectric elements, for example, allows for the generation of droplets at a maximum rate of 3.6 l/min, which is useful for continuous streams. However, these designs require power amplifiers and stable pulses, and droplet size has a nonlinear relationship with pulse width. Therefore, careful tuning and experimentation are necessary to produce consistent droplet sizes. On the other hand, other designs may require different components or techniques, but they offer other advantages, such as low cost or better stability. By exploring and comparing different droplet generator designs, an improved design can be established which is best suited for a specific study.

In order to provide a solution for combustion experiments and other scientific studies, a novel drop on demand generator was designed using of the shelf components. No component manufacturing was required, which resulted in a precise and straightforward device. Single droplets were captured using high-speed imaging. The flow parameters, such as pressure, were controlled by an electronic circuit module along with a microcontroller development board.

3.4 Development of a droplet generator

3.4.1 Construction

Figure 3.10 shows the components of the instrument and Figure 3.11 illustrates the device electrical and hydraulic circuits. The device consisted of six main components: a liquid reservoir, solenoid valve, nozzle, MPX5100DP pressure sensor, Arduino Uno board, and a peristaltic pump. A stepper motor ran the pump, and the pump provided positive displacement flow to the reservoir. The pump type was peristaltic where a force is applied on a tube to push the flow forward. The components, including the pump, are widely available. The pressure sensor provided a feedback signal to the controller. The signal was acquired by the Arduino development board which is based on the microcontroller ATmega328. The microcontroller sampled the sensor analogue signal and provided an output signal to the pump motor, regulating the pressure of the reservoir. Additionally, the Arduino board provided a precise timing control of the solenoid valve. Energising the solenoid allows the flow to pass through the tubing and eventually a droplet was ejected with an initial speed v_i out of a brass nozzle. The nozzles employed in the system were also readily available. Overall, these components enabled the creation of a reliable system that can be assembled, programmed, and operated in a short time.



Figure 3.10: Illustration of the droplet generator components.



(b) Electrical circuit.

Figure 3.11: Hydraulic and electrical circuits of the droplet generator.

Reservoir pressure was controlled electronically, and two methods were employed. First, an ON-OFF controller was implemented. Although this method is considered elementary, the system reliability was shown to be satisfactory. However, this method should be used when slow response and lower accuracy are acceptable. The bang-bang method operates by switching a controlled element when a feedback signal crosses the acceptable error bounds on both sides of a set point. Practically, the microcontroller drove the pump to increase the accumulator pressure up to a setpoint. At that point, the pump was switched off. If the pressure decreased and passed the lower error


Figure 3.12: Block diagram of active pressure control implemented in the controller.

limit, the pump would be operated again. This method was implemented in a loop, repeating the process maintained the pressure approximately at the set point. Furthermore, the second control strategy involved a PID controller. The second method provided responsive pressure compensation. Compared to the ON-OFF method basic implementation, the PID controller resulted in considerable improvement in the time required to achieve a stable output. The PID manual tuning proved essential for stable operation.

Figure 3.12 shows the mathematical representation of the active pressure control implemented in the controller. S(t) represents the pressure setpoint, while e(t) is the difference between the setpoint and the actual pressure P(t). The three parameters K_p , K_I , and K_d are the PID tuneable constants. $\omega(t)$ represents the stepper motor and pump rotational speed. Q(t) represent the flow rate of the system when the valve is operated.

3.4.2 Performance

The device performance was evaluated using pressure response data and droplet dynamics. Figure 3.13 is the time evolution of droplets with increasing injection time. On the figure, the horizontal axis shows the nozzle and ejected droplet resulted from various injection times. Evolution of droplets in time is shown from top to bottom as indicated on the vertical axis. If the injection time is short as indicated at 6 ms of the actual time, a small amount of liquid is observed at the nozzle but retracts soon after and no droplet is ejected. Injection times starting from 2 ms shows droplet formations. The droplets attain a larger size because the ejected stream accumulate when the valve is open. The images at the actual time of 12 ms show that droplets are ejected with increasing initial velocity when injection time is longer. Both actual times of 12 ms and 18 ms indicate that larger droplets can exhibit instability. This instability is due to the inertial forces competing with surface tension. In a later stage, this is dissipated by the viscous forces. Although a droplet might be considered unstable initially, the droplet can reach a stable state if it travels sufficient distance after ejection.



Figure 3.13: Evolution of ejected droplets when injection time is progressively increased. The actual time in the vertical axis shows different frames of the droplet test. Injection time is the time setting of the droplet generator.

3.4.3 Variability

In order to study the variability of the droplet generation process, droplet ejection was repeated five times at the same conditions with a gauge pressure setpoint of 20 kPa. Table 3.2 lists the injection time tested and the equivalent average droplet diameter. It also shows the percentage deviation in droplet diameter and initial speed. Droplet

diameter was measured using images scaled to an object of a known size. The initial speed was obtained using a series of droplet images after detachment from the nozzle, using the speed v = d/t formula, where d is the distance travelled and t is the time duration between images.

Table 3.2 shows that smaller droplets have higher error percentages. Instabilities can affect the measurement, as in the case of 1.67 mm diameter where the deviation was high. Error percentages of the initial speed values are considerably lower than error percentages of the diameter values.

Injection time (ms)	Average diameter (mm)	Max. deviation from average diameter (mm)	Diameter error percentage	Average initial speed (m/s)	Max. deviation from average initial speed (m/s)	Initial speed error percentage
6	0.93	0.1	10.75	0.742	0.024	3.19
8	1.28	0.06	4.69	0.861	0.019	2.17
10	1.43	0.07	4.90	0.981	0.011	1.15
12	1.54	0.07	4.77	1.06	0.012	1.13
14	1.67	0.16	9.58	1.196	0.007	0.56
15	1.76	0.07	4.17	1.206	0.019	1.60

Table 3.2: Droplet diameter and initial speed deviation at setpoint gauge pressure of 20 kPa $\,$

The results of the study on the variability of the droplet generation process are within an acceptable range when compared to other droplet generators mentioned in section 3.3. The percentage deviation in droplet diameter and initial speed are presented in Table 3.2. The table indicates that the smaller droplets have a higher percentage deviation than larger droplets. This could be attributed to the fact that instabilities can have a greater impact on smaller droplets. However, it is worth noting that the percentage deviation of the initial speed values is considerably lower than that of the diameter values, indicating that the generator produces a consistent initial velocity.

Despite the observed deviations, the results obtained in this study are acceptable when compared to other droplet generators. Variability is a common challenge in droplet generator design, and some degree of variation is to be expected. It is important to consider the range of droplet diameters and initial speeds required for specific combustion studies. In addition, steps can be taken to reduce the variability in droplet generation, such as fine-tuning the system or reducing any sources of instability. Overall, the variability results presented in this study are within an acceptable range, and the generator can be considered suitable for use in this study.

3.4.4 Acquisition of pressure sensor readings

The pressure sensor used was MPX5100DP, a gauge sensor with two inlets. One inlet was by design against fluids, as stated in the datasheet. This inlet was connected to the accumulator. The other inlet was left open to the atmosphere. The sensor required no calibration because the additional pressure above atmospheric pressure was measured.

The pressure sensor output was an analogue signal; hence, analogue reader or at least an analogue to digital converter was required. The microcontroller ATmega328 acquired the signal on the Arduino Uno development board. The advantage of this microcontroller is the ability to read analogue signals with 10-bit resolution. In fact, it has five analogue inputs, and only one pin was required in the current system. The sensor value was measured every 100 milliseconds and was used as an input to the control algorithm. The same pressure values were sent in real-time to a computer using the serial interface and USB port available on the board.

3.4.5 Pressure setpoint control and tuning

PID control method was tested for different setpoint profiles implemented in the microcontroller code. This test presented the importance of correct PID tuning. The response could be made faster by removing the differential component of the controller, implementing a PI controller with slight overshoot being tolerated. Flexibility was an advantage in terms of the pressure control strategy. The control method can be chosen to fit other application as required. This feature is not limited to the droplet generator mechanism implemented in this system but can be integrated into other droplet generation techniques. Ultimately, the pressure control pump is suitable for other applications where a particular fluid needs to be maintained at specific conditions.

3.4.6 Control response

The results presented in Figure 3.14 show the response of pressure control using a PID Controller. The PID performed better than the ON-OFF controller, which suffered from fluctuations at the setpoint. The PID method produced a rapid response when changing pressure setpoint. These values were dependent on the parameter tuning in the microcontroller code, sensor capability, and setpoint pressure.

It is evident in Figure 3.14 that the actual signal takes more time to stabilise when the difference between two consecutive setpoints is more considerable. This delay is a direct result of the response time of the instrument. For example, the controller took two seconds approximately to raise the pressure from 3 to 5 kPa; in contrast, it took three seconds to increase the pressure from 5 to 8 kPa.



Figure 3.14: Setpoint signal and pressure response.

3.4.7 Control software

The current droplet generator functions can be significantly enhanced by developing an interface. An application was developed to control the droplet generation process shown in Figure 3.15. The application should allow a fast generation procedure by having a user interface. Parameters such as pressure setpoint can be directly changed when needed, and a custom delay time allows a user to set a specific time to produce a particular droplet size. Droplet stream function can also be integrated to give the ability to eject several droplets with a defined delay between consecutive droplets. Pressure monitoring will allow live monitoring of both the setpoint and the actual measured pressure. Besides, the relation between device settings can be mapped against the ejected droplet properties, hence, enabling the selection of the required size and initial speed of the ejected droplet.

On Demand Droplet									
Stage L Heat Element Lab Conditions T TC-08 T	Connected Disconnected Disconnected Disconnected Disconnected								
Un Demand Droplet Stage Heat Element Lab Conditions TC-08									
Pressure Setpoint (kPa) 8 Set									
Droplet Generation Droplet Stream	Droplet Stream								
1 ms 6 ms Custom Time (ms) 20 Eject Custom Time (ms)									
2 ms 7 ms 40 Eject Delay Time (ms)									
3 ms 8 ms 60 Eject Droplets Count									
4 ms 9 ms 80 Eject									
5 ms 10 ms 100 Eject Start Stop	р								
IO0 Actual Set Point 100 9.3 9.553 40 9.3 9.553 20 Pressure Setpoint (kPa) Actual Pressure	3 3 ure (kPa)								
0	、								
lime									

Figure 3.15: Droplet generator control software showing ability of settings the droplet injection time, pressure, and droplet stream.

3.5 Thermal imaging

Objects with temperatures above absolute zero emit thermal radiation. The maximum thermal radiation emitted by objects is described by Planck's law derived in 1900. The law gives the maximum energy rate an object can emit at a given temperature per unit body surface area per unit wavelength. Maximum energy emission is only achieved by a blackbody which is an idealised object compared to real objects. The ratio between a real body emission at a given temperature to that of a blackbody is known as emissivity. Emissivity of one indicates that a body can absorb all incoming radiation irrespective of radiation wavelength or angle of radiation incidence. Emissivity of one also means that the body can emit the thermal energy given by Plank's law:

$$E_{b\lambda}(\lambda, T) = \frac{c_1}{\lambda^5 \left(e^{\frac{C_2}{\lambda T}} - 1\right)} \,\mathrm{W}\,\mathrm{m}^{-2}\,\mathrm{\mu}\mathrm{m}^{-1}$$
(3.6)

$$c_1 = 2\pi h c_o^2 = 3.742 \times 10^{-16} \text{ W m}^2 = 3.742 \times 10^8 \text{ W } \mu\text{m}^4 \text{ m}^{-2}$$
(3.7)

$$c_2 = \frac{hc_o}{k} = 1.439 \times 10^{-2} \text{ Km} = 1.439 \times 10^4 \text{ Kµm}$$
 (3.8)

$$c_o = 2.998 \times 10^8 \,\,\mathrm{m\,s}^{-1} \tag{3.9}$$

$$h = 6.626 \times 10^{-34} \,\,\mathrm{J\,s} \tag{3.10}$$

$$k = 1.381 \times 10^{-23} \text{ J K}^{-1} \tag{3.11}$$

where $E_{b\lambda}(\lambda, T)$ is called spectral blackbody emissive power, λ is the wavelength (µm), T is the abosulte body temperature (K), C_1 is the first radiation constant, C_2 is the second radiation constant, c_o is the speed of light in vacuum, h is Planck's constant, and k is Boltzmann's constant.



Figure 3.16: Spectral blackbody emissive power at several temperatures and operating infrared bands of common detectors.

The emitted radiation increases considerably with temperature and the maximum spectral emissive power shifts towards shorter wavelengths. When the temperature becomes high enough, objects start to glow in red colour and then progressively move toward brighter colours as more of the radiation shifts towards the shorter wavelengths in the visual spectrum.

Figure 3.16 illustrates the spectral blackbody emissive power at various temperatures. The graph demonstrates how the intensity of emitted radiation by a blackbody changes with respect to wavelength for different temperatures. As the temperature of the blackbody increases, the peak of the curve, which represents the wavelength with the highest emissive power, shifts toward shorter wavelengths.

Integrating Plank's law of radiation over all wavelengths leads to Stefan-Boltzmann law which allows the calculation of the total blackbody emissive power E_b :

$$E_b = \int_0^\infty E_{b\lambda}(\lambda, T) d\lambda = \sigma T^4 \text{ W m}^{-2}$$
(3.12)

$$\sigma = \frac{\pi^4 C_1}{15 C_2^4} = 5.670 \times 10^{-8} \,\mathrm{W \,m^{-2} \,K^{-4}}$$
(3.13)

where σ is called Stefan-Boltzmann constant.

When taking infrared thermal measurements, cameras detect only a limited band of the whole spectrum, integrating Planck's law in a certain wavelength band $\lambda_1 - \lambda_2$ enables the evaluation of energy flux detected by a sensor in its operating band:

$$E_{b,\lambda_1-\lambda_2}(T) = \int_{\lambda_1}^{\lambda_2} E_{b\lambda}(\lambda, T) d\lambda$$
(3.14)

Evaluating the energy flux fraction detected by middle wavelength infrared (MWIR) and long wavelength infrared (LWIR) cameras can show their respective advantage over the other. MWIR and LWIR bands are 3-5 µm and 8-12 µm, respectively. A 300 K blackbody emits 1.6% of its total radiation in the MWIR band, and 26% in the LWIR band. Clearly, the LWIR is the suitable option for this temperature. However, when the blackbody temperature is increased to 600 K, the MWIR becomes a suitable option just as LWIR with radiation fractions of 23% and 21%, respectively. The radiation fraction is not the only factor that affects the operating band choice. Other factors include the object features such as emissivity and environment optical properties.

3.5.1 Real body radiation

The radiation of real bodies is different from that of a blackbody. Blackbody emission and absorption represent the upper limit that may be approached by real bodies. This limit can be approached under certain conditions and certain wavelengths. The incident radiation on a real body is partially absorbed, unlike a blackbody on which the radiation is completely absorbed. The rest of the radiation may be partially transmitted through the real body and/or reflected. Donating the absorbed irradiation fraction by α_r , reflected irradiation by ρ_r , and transmitted irradiation by τ_r , the following relation applies:

$$\alpha_r + \rho_r + \tau_r = 1 \tag{3.15}$$

where α_r , ρ_r , and τ_r are also known as absorptivity, reflectivity, and transmissivity coefficients.

Figure 3.17 illustrates the reflection, absorption, and transmission of incident radiation by a semi-transparent medium. The diagram shows that when incident radiation encounters the medium, it can undergo three different interactions: reflection, absorption, or transmission. The reflected portion of the radiation bounces back off the surface of the medium without entering it. The absorbed part of the radiation is taken in by the medium and converted into other forms of energy, such as heat. Lastly, the transmitted portion of the radiation passes through the medium and emerges on the other side. The sum of these three interactions equals the total incident radiation, highlighting the fact that energy is conserved throughout these processes. The properties of the semi-transparent medium and the characteristics of the incident radiation, such as wavelength and angle of incidence, determine the relative proportions of reflection, absorption, and transmission.



Figure 3.17: The reflection, absorption, transmission of incident radiation by a semitransparent medium.

In absorbing mediums such as air, the transmissivity coefficient is exponentially related to the travelling distance L of radiation:

$$\tau_{\lambda} = e^{-\zeta_{\lambda}L} \tag{3.16}$$

where ζ_{λ} is the spectral absorption coefficient (m⁻¹).

A real body emits only a part E of the radiation E_b emitted by a blackbody at the same temperature. The emissivity is the ratio between a real body radiation to that of a blackbody

$$\varepsilon_t(T) = \frac{E(T)}{E_b(T)} \tag{3.17}$$

where $\varepsilon_t(T)$ is the total hemispherical emissivity.

The emissivity can also depend on the wavelength $\varepsilon_{\lambda}(\lambda, T)$ and direction. A grey surface has properties independent of wavelength, and a diffuse surface has properties independent of direction.

Given that a real surface may not be truly grey over all the spectrum, often a real body can have a constant emissivity in the used infrared sensor band. Therefore, the grey surface assumption can be satisfied.

The emissivity and absorption coefficients are related by Kirchhoff's law:

$$\varepsilon(T) = \alpha(T) \tag{3.18}$$

For opaque surfaces $(\tau_r = 0)$ and by applying Kirchhoff's law, the emissivity is linked to the reflectivity:

$$\varepsilon_{\lambda} + \rho_{r\lambda} = 1 \tag{3.19}$$

Consequently, materials with high emissivity reflect a small fraction of the radiation coming from the surroundings. While shiny materials like metals, which have low emissivity, reflect a large amount of the background radiation.

3.5.2 Calibration

Calibration of a thermal camera is essential to ensure accurate and reliable temperature measurements over time. While cameras typically come with factory calibration, several factors can cause measurement drift or inaccuracies, including aging components, environmental conditions, or other physical changes, such as the use of different lenses or filters. Lenses and filters can alter the camera's field of view and spectral response, potentially affecting temperature measurements. Regular calibration ensures that the camera remains accurate and maintains optimal performance, taking into account the specific operating conditions, emissivity, atmospheric transmissivity, and the impact of lenses and filters. Additionally, calibration helps identify and correct any discrepancies or errors that may arise from changes in the camera's hardware or software.

The output of infrared detectors depends on the received radiation energy and calibration is needed so that the output can be converted into the target object temperature [188]. Figure 3.18 indicates that the radiation of the target object $\varepsilon E_{b,obj}$ is only a part of the radiation detected by the infrared camera where ε is the emissivity of the target object. The target surface temperature is T_{obj} . The surrounding ambient environment is assumed to have a constant temperature T_a and its radiation is reflected by the target surface $(1 - \varepsilon)E_{b,a}$ where the coefficient $(1 - \varepsilon)$ represent the reflectivity according to equation 3.19. The atmosphere, which has a temperature of T_{atm} , contributes to the received radiation $(1 - \tau)E_{b,atm}$ where τ is the transmitivity of the atmosphere. The atmosphere also attenuates the radiation originating from the target object $\varepsilon \tau E_{b,obj}$ and the reflected ambient radiation $(1 - \varepsilon)\tau E_{b,a}$ and that is the reason for including τ in the mentioned terms. Furthermore, the camera is assumed to compensate for its radiation internally. In the context of thermal radiation measurement, ambient temperature and atmospheric temperature refer to two distinct concepts that impact the measurements.

Ambient temperature refers to the temperature of the surrounding environment in which a measurement device, such as a thermal camera or radiometer, is operating. This temperature can influence the accuracy and performance of the device, as it may introduce errors due to temperature drift or changes in the device's sensitivity. Therefore, it is essential to consider the ambient temperature when calibrating the instrument and interpreting the measurements.

Atmospheric temperature, on the other hand, relates to the temperature of the air or gases present in the path between the measurement device and the object being measured. In the case of thermal radiation, the atmosphere can absorb, emit, or scatter some of the thermal radiation that travels between the object and the measuring instrument. This can lead to inaccuracies in the measurement, as the detected radiation may not accurately represent the object's true temperature. To account for this, corrections must be made based on the atmospheric temperature, the distance between the object and the instrument, and the composition of the atmosphere.

In summary, ambient temperature affects the performance and accuracy of the measurement device, while atmospheric temperature influences the thermal radiation's interaction with the surrounding atmosphere, potentially altering the detected radiation and subsequent temperature readings.



Figure 3.18: Illustration of radiation detected by infrared camera.

Summing up the three terms, the total energy flux E_t detected by the infrared camera:

$$E_t = \varepsilon_t \tau_r E_{b,obj} + (1 - \varepsilon_t) \tau_r E_{b,a} + (1 - \tau_r) E_{b,atm}$$
(3.20)

Given that the camera temperature is compensated, the output signal of the camera U has a linear relation with the collected radiation E_t :

$$U = \varepsilon_t \tau_r U_{obj} + (1 - \varepsilon_t) \tau_r U_a + (1 - \tau_r) U_{atm}$$
(3.21)

The main step of the calibration process is to approximate the camera output with a function similar to the Plank's law. Many approximation have been evaluated against their quality to fit the output response [189], [190]. One of such approximations is the Sakuma–Hattori equation for narrow band infrared detectors [190], [191]:

$$U = \frac{C}{\exp\left(\frac{c_2}{AT+B}\right) - 1} \tag{3.22}$$

where T is the temperature in kelvins, c_2 is the second radiation constant, and A, B, and C are fitting parameters. The parameters are a function of integration time and the imaging system characteristics [192]. By substituting equation 3.22 in equation 3.21, the temperature of the object can be written as:

$$T_{obj} = \frac{c_2}{A \ln\left(\frac{\varepsilon_t \tau_r C}{U - (1 - \varepsilon_t)\tau_r U_a - (1 - \tau_r)U_{atm}} + 1\right)} - \frac{B}{A}$$
(3.23)

Equation 3.23 shows that evaluating the object temperature requires the ambient background and atmospheric temperatures which usually coincide, emissivity of the target object, the atmosphere transmissivity, and the fitting parameters. A blackbody oven with emissivity very close to 1 can be used to find the three fitting parameters. Furthermore, atmospheric transmissivity is practically equal to 1 if the oven is relatively near the camera. Therefore, equation 3.21 can be simplified:

$$T_{obj} = \frac{c_2}{A\ln\left(\frac{C}{U}+1\right)} - \frac{B}{A}$$
(3.24)

By varying the blackbody temperature over the range of interest, several camera outputs can be recorded. The data is then used in an optimisation procedure to find the best-fit parameters. Equation 3.24 or 3.22 is solved for the camera output directly as shown in Figure 3.19 for for Hamamatsu C12741-03 InGaAs camera.



Figure 3.19: Calibration curve example with measured points U in counts against a blackbody temperature for Hamamatsu C12741-03 InGaAs camera.

3.6 Conclusion

In this study, several experimental setups have been developed and implemented to investigate droplet evaporation, droplet surface tension, and the interactions of droplets with burning wood surfaces. Four distinct setups are employed: the first to examine the effects of particles on droplet evaporation; the second to investigate the influence of particles on droplet surface tension; the third to explore the phenomenological aspects of droplet impact on burning wood surfaces; and the fourth to characterise the thermal and surface properties of wood samples affected by a single water droplet.

The imaging methods utilised in these setups can be divided into two categories: visual and thermal techniques. Visual approaches involve the use of cameras to track droplet evaporation and document the spreading of droplets when they come into contact with burning surfaces. In addition, visual methods include surface mapping techniques based on focus variation. Thermal techniques, on the other hand, make use of thermal cameras and sensors to analyse the thermal properties of surfaces. This chapter presents a general overview of these techniques, with subsequent chapters offering more detailed descriptions of each method and their corresponding experimental results.

Chapter 4

Evaporative and Interfacial Characteristics of a Droplet with Suspended Particles

An experimental investigation was conducted on water droplet surface contaminated by smoke particles, which is most likely in a real firefighting scenario. Droplets were evaporated by external heating to show the effect on one of the primary four mechanisms of fire extinguishment. Mechanisms of fire extinguishment are all affected by the interaction of water spray and fire, which starts with water droplets passing through a smoke layer. Unlike previous experimental investigations on water interaction with fire conducted in large scale setups, chambers, and real scale rooms, this work demonstrates the significance of smoke contaminants on water droplet evolution and surface tension. Quantitative measurements on evaporating rate showed that contaminated droplets had a higher rate of evaporation. The results were consistent with available research on nanoparticle effect in fluids. However, unlike the previous experimental research which investigated the change of properties by volume mixing while this study focused on surface contamination as this is most likely in real situations. Furthermore, a theoretical analysis of effective thermal conductivity is given and compared to that of Maxwell theory in Chapter 5. Additionally, the surface tension, which is of great importance to spray characteristics, was found to decrease by almost a factor of two for the surface contaminated droplets. The expected behaviour when droplets become considerably small is discussed. Surface tension change can influence spray characteristics considerably in fire extinguishment systems.

4.1 Introduction

An experimental investigation was conducted on water droplet surface contaminated by smoke particles. Droplets were evaporated by external heating to show the effect on one of the primary four mechanisms of fire extinguishment. When water is sprayed over a fire plume, it serves to extinguish it by various mechanisms. Primary mechanisms are heat extraction and air displacement [48]. Heat is extracted from flames or the combustible surface by droplet evaporation. The water vapour produced from the evaporation displaces air and reduces oxygen concentration. These mechanisms are all affected by the interaction of water spray and fire, which starts with water droplets passing through a smoke layer. Experimental investigations on water interaction with fire have been conducted in large scale setups, chambers, and real scale rooms in the past. These experiments mainly focused on the suppressing efficiency of water spray [48], [193]. In practice, water spray is contaminated by smoke particles during fire quenching. The main aim of this chapter is to show the significant impact of smoke contaminants on the evaporation process as well as the surface tension of water droplets.

Smoke particles have been previously investigated in terms of their size distribution and composition. Studies were focused on both smoke from application-based burning and fire tests. Kleeman and others [24] found that smoke produced by wood contained not only carbon particles but also a variety of organic compounds and metal particles. Similar results were obtained when analysing boiler smoke [194]. Moreover, smouldering in masonry heaters produces fine smoke particles [25]. The studies mentioned above have all agreed that the produced smoke particles were below 1µm and peaks at 100nm approximately.

Previous experimental studies on smoke particles of wildfire and test fires are summarised in table 4.1. It is noticeable that smoke which was analysed immediately after production showed a peak at 100 nm. While transported smoke, which tends to agglomerate, showed peaks at larger sizes. Furthermore, smoke particles vary in their chemical composition. Although the composition depends on their source, carbon soot was found to be the main predominant agglomerate in wood combustion [24], [25], [26]. In addition, smoke particles can also contain metals such as potassium, iron, magnesium, sodium, and copper. Other materials such as silicon, phosphorus, and sulphates can be detected as well. Due to the size and composition of smoke particles, it is expected that surface contaminated droplets have different surface behaviours.

Combustibles	Measurement environment	Test range	Peak value	Reference
Wildfire	Transported smokes	10 nm - 10 µm	$300\mathrm{nm},600\mathrm{nm}$	[26]
Timber harvest residues	Forest	10 nm - 43 µm	$150\mathrm{nm}$	[143]
Space vehicles common materials	Microgravity test on ISS	20 nm - 1 µm	$100{\rm nm}$ - $300{\rm nm}$	[45]
Buildings common materials	Small scale cone calorimeter	40 nm - 10 µm	No exact peak	[195]
Polymers	Small lab scale	1 μm - 10 μm	$2\mu{ m m}$	[144]
Woods	Medium Lab scale	$6\mathrm{nm}$ - $500\mathrm{nm}$	$10{\rm nm}$ - $100{\rm nm}$	[196]
Wildfire	Transported smokes	30 nm - 1 µm	$100\mathrm{nm},500\mathrm{nm}$	[145]
Wildfire	Airborne measurements	10 nm - 1 µm	$50\mathrm{nm}$ - $150\mathrm{nm}$	[197]
Wood	Small lab scale	$20\mathrm{nm}$ - $10\mathrm{\mu m}$	$20\mathrm{nm},300\mathrm{nm}$	[146]

Table 4.1: Size properties of smoke particles in wildfires and test fires

Rasid and Zhang [27] have examined surface contamination in a fuel droplet and reported the influence of nanoparticles on thermo-physical properties, they found the surface soot contamination has significantly altered the diesel droplet's burning characteristics. They showed that the evaporation and burning rates of the droplets were reduced and that the stability was critically affected. However, they focused on the burning behaviour of the droplets without further experiments on the evaporation behaviour as an individual process.

Certainly, the problem is complex due to the variables of smoke content, particle size distribution, particle agglomeration, chemicals reaction with water, and the type of forces that arise between water and contamination. Considerable research has been conducted on smoke particles, the interaction between fire and water spray, and nanoparticle effect on fluids. However, no experimental investigation on the fundamental properties of smoke particle contaminated water droplets has been conducted so far. Therefore, this experimental analysis aims to investigate the influence on evaporation rate and surface tension of water droplets. Water spray has a significantly larger surface area than that of a single droplet having a comparable mass. The spray droplet size distribution is critically dependent on properties such as the surface tension. Knowing the smoke effect on the droplet's surface tension is of importance because most droplets will further break up in a turbulent flow besides evaporation. Although the possibility of contamination is much higher in real applications compared to a single droplet test, studying a single droplet system enables enhanced magnified imaging throughout the lifetime of the droplet.

4.2 Experimental apparatus

Figure 3.5 shows the experimental setup. The setup was placed in an average temperature and humidity condition of 23°C and 38%, respectively. Water droplets were injected using a 1 mL syringe and suspended on 100 µm silicon carbide fibre. The initial diameter of droplets was 1 ± 0.1 mm for all tests. The water used in this experiment was distilled water. To track the lifetime of an individual droplet, a Phantom SA4 was used with Nikon AF Micro 60 mm f/2.8 lens. In addition, backlighting of droplets was achieved by IDT LED intense illuminator and a diffuser.

A preliminary case study was performed to show the effect of pine wood smoke particles on water droplet evaporation. The goal was to apply particles on water droplets of 1 mm in diameter using smoke from small wooden pieces, as shown in Figure 4.1 (a). Smoke was applied for a duration of five seconds. The concentration of the smoke particles on the surface of the droplet was not measured due to the setup limitation. The initial droplet size was monitored by the camera live view to ensure that the size was within a tolerable range. The droplets were then exposed to a heating element, as shown in Figure 4.1 (b).



Figure 4.1: a) Suspension of a water droplet by a fibre and applying smoke particles on its surface; b) Droplet evaporation by a heating element; c) Temperature measurements of at a point in the flow field created by the heating element. The distance between the wood pieces, heat element, and droplet was 10 mm.

Figure 4.1 (c) indicates how the temperature was measured at the same location where the droplet was suspended. A K-type thermocouple was used with a Pico TC-08 Data Logger to record the temperature. The thermocouple was located at the same point as the droplet suspension point. The time evolution of air flow temperature that was produced as a result of the heating source is shown on Figure 4.2. The entire evaporation process of each droplet was captured. This was then compared to the distilled water droplet evaporation without applying smoke particles. This study provided an initial indication on the lowest amounts of contamination that can affect evaporation and surface tension of a water droplet.



Figure 4.2: Temperature measurements of the flow created by the heating element at voltage 4 V voltage and a distance of 10 mm above the heating element.

The droplets were evaporated using a constantly powered heating source. The source was designed to keep its power constant during evaporation. In each test, the process started by supplying the same power to a stainless-steel heating element. The element is then allowed to reach a steady state. Once the steady state is reached, the coil is moved under the droplet by a linear actuator. The actuator ensured the consistency of heating element placement under the droplet.

Droplet evolution images were processed using a MATLAB script. The algorithm used area segmentation methods to calculate droplet area, diameter, and centroid. In addition, droplets were assumed to be spherical based on the two-dimensional shape captured by the camera. Using this data, droplet normalised diameter reduction $(D/D_o)^2$ with time t/D_o^2 was evaluated according to D^2 -law. The derivation of dsquared law is detailed in Appendix A. The mass loss rate which leads to this law was used previously in several studies related to water evaporation. For example, droplet evaporation was investigated under different conditions, such as the hot air effect in fire extinguishment [10]. The law was used to illustrate the enhancement of fire suppressions by radiation attenuation [85]. It is used to highlight the effect of smoke particles on a water droplet surface.

4.3 Preliminary results

Figure 4.3 shows evaporation of distilled water droplets and Figure 4.4 presents the evaporation of droplets with smoke particles. The results indicated three stages, starting with a heating period where the curve slope gradually increases, followed by a second stage where the slope is relatively steady, and ending with the final stage where there are disturbances to the shape of the droplet.



Figure 4.3: Evaporation of distilled water droplets. The Vertical axis represents the square of normalised diameter while the horizontal axis is time scaled by initial diameter.

Quantitative measurements were only made on the second stable stage. The mean evaporation rate was 0.069, $0.064 \ s/mm^2$ for contaminated and pure surface respectively. Coefficients of variance were 4% and 9%, respectively. The contaminated surface enhanced the evaporation rate by 8%. It was observed in this study that the particles on the surface were not agglomerated but rather distributed. It is noticeable that this effect is arising from surface contamination only, while the effect described in the literature was based on volume mixing of different particles and fluids. A similar effect has been reported in previous studies on nanoparticles [198]. The explanation

was that the enthalpy of vaporisation and surface tension were reduced when adding specific particles to the bulk of a fluid. Hence, a higher evaporation rate.



Figure 4.4: Evaporation of water droplets with surface suspended with smoke particles.

Previous studies on the topic of nanoparticles on base fluids have shown enhancements of heat transfer. Xuan et al. [199] measured the increase in thermal conductivity with added copper nanoparticles up to 7.5% in volume concentration. In Kakac's review of thermal conductivity enhancement with nanofluids [200], theoretical work was firstly conducted by Maxwell [201] and then followed by other researchers to develop the models progressively. These models assume a discontinuous bulk phase. Furthermore, the review covered the theoretical and experimental work on the increase of convective heat transfer due to effective thermal conductivity and Brownian motion. Previous research deals with properties enhancement as it originates from the volume mixing of nanoparticles in a base fluid. Nevertheless, water used in fire extinguishment is surface contaminated with smoke particles rather than bulk mixed. Hence, it is expected that the surface behaviour of water droplets changes when it is contaminated. Although the increase in the evaporation rate was relatively small in preliminary results, a low amount of contamination has a major impact on surface tension.

4.3.1 Optical properties

Radiation attenuation is an indirect method by which water spray absorbs radiant heat. Radiation absorption in water occurs as a volumetric phenomenon rather than a surface process only because water is semi-transparent [85]. In the case of this experiment, surface appearance was significantly altered, and this was an indication that properties such as emissivity were affected. Further experiments are suggested in order to understand the optical contaminated droplet properties and radiation contribution.

Additional experiments are necessary to gain a deeper understanding of the optical properties of contaminated droplets and the role of radiation in this context. In relation to the method of fire extinguishment using a water curtain, these experiments will help elucidate the effectiveness of the water curtain in absorbing and scattering radiant heat from the fire. By examining the behaviour of contaminated droplets and their interaction with radiation, researchers can potentially optimise the design and deployment of water curtains in firefighting scenarios. This could lead to improved fire suppression techniques, increased safety for firefighters, and a reduction in property damage caused by fires. A comprehensive understanding of the underlying mechanisms of contaminated droplets and radiation will ultimately contribute to the development of more efficient and effective fire extinguishment strategies.

4.3.2 Surface characteristics

Surface tension influences liquid transport and heat transfer phenomena. It is generally understood that droplets on fibre take different shapes depending on the characteristics of droplets, fibre, surrounding environment, and surface interaction between them. Examples of these shapes are barrel, clamshell, hanging by a triple line, and other configurations based on the stability of the droplet [202]. Moreover, a droplet on fibre configuration is affected by gravity and the droplet tend to be elongated. Elise et al. [203] have studied capturing droplets on thin horizontal fibres. Their analysis provides a method to measure surface tension for a barrel shaped droplet which is the same configuration in this experiment. Figure 4.5 shows the geometric parameters needed to evaluate the surface tension. Figure 4.6 is a plot of sin α against R^3 for pure and surface contaminated droplets; where α is the angle between the horizontal through the centroid and fibre first point of contact with droplet surface, R is droplet radius. The Figure shows a linear relationship approximately. Assuming the bulk density of water is 997 kg m⁻³ and gravity acceleration is 9.81 m s^{-2} . Based on the linear regression of the data, it was found that surface tension values were 50.7 and 30.3 mN m^{-1} for pure droplets and contaminated droplets, respectively. The coefficients of variance were 4% for both Cases. Interestingly, although the concentration of smoke particles increased with evaporation, the linear relation in Figure 4.6 remains constant.



Figure 4.5: A single droplet suspended by a fibre showing droplet centroid, radius, angle between the horizontal axis through the centroid and the fibre, and fibre diameter.



Figure 4.6: Values of $\sin \alpha$ versus cubed droplet radius.

Surface tension is inversely proportional to Weber number which is given by equation 4.1.

$$We = \frac{\rho v^2 l}{\sigma} \tag{4.1}$$

where ρ is the liquid density, v is the velocity, l is the characteristic length, typically a droplet diameter and σ is the surface tension. Weber number influences spray characteristics, representing the ratio of disruptive hydrodynamic forces to the stabilising surface tension force. Low Weber numbers indicate the fluid surface energy is dominant, and droplets tendency for splashing is lower. In contrast, high Weber numbers describe high kinetic energy and that spray drops are less stable and easy to break up. Changes to Weber number would influence the mechanism by which water droplets break up in fire extinguishment systems [204]. For Example, Splashing and bouncing are considered to be limiting factors for drop deposition efficiency on surfaces [205]. Furthermore, the introduction of water mist systems led to the requirement for high speed droplet velocities to increase plume penetration and achieve surface impingements [206]. These processes are critically dependant on Weber number, and the effect presented in this work shows that surface contamination can play a significant role in droplet breakup, splashing and impingement characteristics. The trends in this study are consistent with the available research [149], [207]. A similar effect has been realised in evaporating pendant droplets [133], but some particles showed no effect on surface tension. On the other hand, it has been shown that surface tension can be increased when the concentration of nanoparticles increases [208]. On a molecular level, smoke particles could behave as hydrophobic nanoparticles. When wet hydrophobic particles become adsorbed at surfaces, it leads to intermolecular separation of water molecules. The attraction of water molecules is lowered, leading to a reduction of surface tension [209].

Literature results showing the effect of particles on surface tension may seem to have many contradictions. However, it's worth noting that the behaviour of particles at the interface of a liquid can be highly dependent on factors such as particle size, shape, and surface properties, as well as the properties of the liquid itself. Some studies may have used particles with different properties or investigated different types of liquids, leading to different results. Additionally, the methods used to measure surface tension can also have an impact on the observed behaviour.

Moreover, a closely related concept is the Marangoni effect, where the mass transfer occurs along surfaces induced by surface tension gradient. Surface tension gradients are usually induced by temperature variations and have a negligible effect on evaporation [210]. By observing contaminated droplet images as shown in Figure 4.7, it can be suggested that the droplet's surface becomes uneven in terms of smoke particles distribution. This unevenness would influence the surface tension gradient on the droplet surface and enhance surface circulation and Marangoni effect.

When the size of droplets becomes similar to the mean free path of the surrounding air, the evaporation is limited by the transport rate of the vapour layer on the surface of droplets. Theoretical analysis of this has been established by Fuchs [105] where the diffusion process does not occur on the surface of a droplet but rather at a distance equivalent to the mean free path on the surface. This effect is not important for millimetre droplets; however, it is of great significance for micrometre sized droplets. Essentially this is due to substantially few molecules being available in the latter case.

Considering a very small droplet, the evaporation rate is different because of the increased vapour pressure of small droplets. The curvature of droplets and their small size means that they have a higher vapour pressure compared to flat surfaces. Kelvin equation shows the relation of vapour pressure to droplet size and surface tension:

$$\ln \frac{P}{P_{\rm sat}} = \frac{2\gamma M}{r\rho_L RT} \tag{4.2}$$



Figure 4.7: Time evolution of evaporating contaminated droplet showing uneven surface.

where P (Pa) is the vapour pressure at the droplet surface, P_{sat} (Pa) is the saturated vapour pressure on a flat surface, γ (N/m) is the interfacial tension, M (g/mol) is the molecular weight, r (m) is the droplet radius ρ_L (kg/m³) is the liquid density, R (J/(mol · K) is the universal gas constant, and T (K) is the temperature.

As r increases, vapour pressure P decreases and tends to the saturation vapour pressure. The curvature becomes smaller, and droplets grow to achieve similar behaviour to that of flat surfaces. For example, for a pure droplet of 1 mm in size with a density of 997 kg/m^3 at a temperature of 25 °C under atmospheric pressure 101.325 kPa. Given that surface tension is 72 mN/m and the vapour gas constant is 461.52 J/(kg K), the effect is negligible. However, for droplets of 1 µm and 0.1 µm in size, the vapour pressure increases by 0.2% and 2%, respectively. Furthermore, a decrease in surface tension will lead to a decrease of the surface vapour pressure according to Kelvin equation. Therefore, if a similar reduction in surface tension, as shown above, occurs for smaller droplets, vapour pressure might experience two competing mechanisms for micrometre and sub-micrometre droplets. Recently, researchers found that Kelvin equation was accurate to the nanometre scale [211].

4.4 Notes on initial investigations

Water droplets evaporation has been shown to be affected by smoke particles contamination. In particular, the surface of water droplets was deposited with smoke particles to simulate the effect of using water in an actual fire extinguishment situation. Quantitative measurements on evaporating rate showed that contaminated droplets have a higher rate of evaporation. The results were consistent with available research on nanoparticle effect in fluids. However, previous experimental research investigates the properties changes by volume mixing while this study focused on surface contamination, which is most likely in a water spray fire suppression process. Additionally, the surface tension was found to be lower for surface contaminated droplets. Surface tension change can influence spray characteristics considerably in fire extinguishment processes.

The next sections detail an improved setup for droplet evaporation and surface tension measurements. The improved setup allowed for droplet temperature measurements using a fine thermocouple rather than a fibre. In addition, pendant drop tensiometry enhanced the results and enabled dynamic variation of droplet volume for further analysis.

4.5 Improved experimental apparatus

Figure 4.8 is schematic of a smoker used to apply smoke particles on droplets. The smoker works by directing a fresh air flow from the surroundings that passes around a burning piece of wood by means of an impeller. The flow carries the smoke emerging from the wood which is then directed towards a suspended droplet. The distance between the flow outlet and the water droplet was 7 cm. The speed of the flow at the outlet was $4.6-4.9 \text{ m s}^{-1}$ while the speed at the droplet location was $0.5-0.8 \text{ m s}^{-1}$. The temperature of the flow at the location of the droplet was that of room temperature because of the cooling effect of the flow intake from the room.



Figure 4.8: A schematic of the smoker used to apply particles on droplet surface buy directing a flow passing around a burning wood sample.

The thermocouple wire was relatively thin with a diameter of 0.075 mm. This wire is prone to thermal expansion. Since the camera and lens setup was zoomed on the droplet with a thin depth of field, the thermocouple wire was required to keep its position at the focus point. Therefore, a tension mechanism with flexible links was designed as shown in Figure 4.9. The flexible joints work as springs when bent. Specifically if they are bent towards each other, an opposite force will act and push the links towards their initial position. The thermocouple wires were wrapped around the edges of the flexible links which push the links towards each other. The wires were adhered to the links by a tape to prevent the release of the wires around the edges. The thermocouple in this situation stayed stretched even when it experienced thermal expansion.



Figure 4.9: An attachment with flexible joint for thermocouple wire stretching.

Figure 4.10 shows an improved setup for single droplet evaporation. The setup features a vertical and horizontal motorised stages for accurate position of the droplet relative to the heating element. A micrometer is also used for moving the droplet towards or away from the camera lens. This micrometer was used to improve the focus and to overcome the narrow field of depth resulted from the imaging setup. The heating element was actuated by a pneumatic cylinder which had a stroke of 10 cm. The light was used along with a diffuser made out of a white acrylic sheet. The stage motors, cylinder, and camera trigger were controlled using an application developed for this setup.



Figure 4.10: Improved single droplet evaporation setup featuring motorised axes, focus micrometer, thermocouple stretching attachment, and a heating element pneumatic cylinder.

4.6 Results and discussion

Figure 4.11 and Figure 4.12 show the evaporation of a pure droplet with an average gas temperature of $T_a = 23$ °C and $T_a = 304$ °C, respectively. Figures 4.13, 4.14, and 4.15 show the evaporation of water droplets with smoke particles applied for 10, 120, and 240 seconds. Along with the evaporation curve in the figures, the gradient of a 10 piecewise linear fit of the evaporation curve is presented. The gradient is divided into 10 sections, and then each section is fitted with a linear model to visualise and compare the trends in the gradient.



Figure 4.11: Pure droplet evaporation at $T_a = {}^{\circ}C$.



Figure 4.12: Pure droplet evaporation at $T_a = 304$ °C.



Figure 4.13: Evaporation of a droplet applied with smoke particles for 10 s at $T_a = 304$ °C.



Figure 4.14: Evaporation of a droplet applied with smoke particles for 120 s at $T_a = 304$ °C.



Figure 4.15: Evaporation of a droplet applied with smoke particles for 240 s at $T_a = 304$ °C.

The gradient clearly show that the first region has a higher value. This is because of the transition from the droplet heating, where the evaporation is very minimal, to the evaporation stage. The evaporation constant was measured based on the middle 8 regions. The evaporation constant is defined in Appendix A.

The results of different smoke application times do not indicate a major change in the evaporation constant. The results are presented in Figure 4.16. The pure droplets had the minimum evaporation constant of $0.0569 \text{ mm}^2 \text{ s}^{-1}$, while droplets with smoke particles had 0.0589, 0.0613, 0.0618 mm² s⁻¹ for smoke application times 10, 120, 240 seconds, respectively. The trend from the plots indicate that the evaporation constant increases as smoke particle application time increases. The percentage change between pure droplets and droplets with particles was 3.5%, 7.7%, and 8.6% for times 10, 120, 240 seconds, respectively.

Figures 4.12 to 4.15 indicate that the last stage of evaporation in all cases experienced lower evaporation constant compared to the major part of the droplet lifetime. Figure 4.17 presents the averaged last stage droplet evaporation constants.


Figure 4.16: Evaporation constant of a droplet applied with smoke particles for different times at $T_a = 304$ °C. The experiment was repeated 10 times at each smoke application time.



Figure 4.17: Last stage evaporation constant of droplet applied with smoke particles for different times at $T_a = 304$ °C. The experiment was repeated 10 times at each smoke application time. The ANOVA test shows that the F value is greater than F critical. The ANOVA test indicates that the results of different application times are significant.

4.7 Measurements of surface tension

This work aims to demonstrate the effect of particles on the surface tension of water drops. Prior to the discussion of the surface tension relevance in this work, a definition of its physical meaning is presented. Surface tension can be defined by considering the molecular and mechanical nature of fluids. The molecular level is covered by the theory of Bakker, while the mechanical definition is presented by Laplace equation [159].

Two immiscible fluids, such as air and water, brought together are considered here. Sometimes, the surface tension is explained as follows. In the bulk of water, a molecule is surrounded by many other molecules that attract each other. These forces are considered symmetrical around this molecule, leading to zero net force acting on it. But close to the surface, the attraction forces are no longer symmetrical because the forces near the surface are less compared to that in the bulk of water. Consequently, a net force is directed towards the bulk of the medium. This view of the problem is wrong. Indeed if a resultant force acted on the molecule, it would be accelerated towards the bulk. Hence, the molecule close to the surface will arrange themselves in a way that the resultant force in the normal direction to the surface is zero. This also implies that the pressure gradient in the same normal direction is zero.

The normal pressure and the pressure parallel to the surface acting on a molecule in the bulk are equal. However, they are not equal close to the surface. Therefore, the tangential pressure is a function of the distance from the surface. The tangential pressure difference between near the surface and the bulk is what contracts the surface and creates a tension per unit length. The boundary between the two mediums is not a plane, but rather, it is a transition layer between the two mediums. The mechanical nature of this boundary is represented by a hypothetical stretched film.

Using surfactants to control the surface behaviour of liquids is based on the principle of weakening molecular interactions at interfaces. In contrast, researchers believe that nanoparticles affect surface tension by generating an opposing pressure to the surface tension of the base fluid [205]. The nature of these particles means that surface tension can be altered by particles' size [212], wetting, packing structure **Huerre2018**, and pacing between particles **Garbin2012**.

Previously, attachments of particles to a bubble surface in air-liquid systems were attempted using mechanical stirring. This method enables the particles to be applied during the measurement of interfacial tension using the maximum bubble pressure method. Particles can form close-packed structures at interfaces. Particles can be affected by the flow near the interface, which might shift away the surface from equilibrium **Huerre2018**. Thus, the maximum pressure method can be a limiting factor when using stirring techniques.

Surfaces covered by nanoparticles take significantly long time for adsoption when used with maximum bubble method. Sessile and pendant drop tensionmetry have been shown to be reliable in such cases, including situation when surfaces experience expansion and contraction **Garbin2012**. The applicability of different methods was discussed in Chapter 3.

4.7.1 Stability of droplets

The volume of a droplet is very critical to the pendant droplet method. The measurement technique sensitivity can be investigated by forming a range of drops with different needle sizes and Bond numbers. The liquid considered is clean distilled water droplets with a known surface tension of 72.6 mN m^{-1} and density of 1000 kg m^{-3} .

Experimental results revealed that droplets become unstable near the maximum attainable volume for a specific needle size. Furthermore, the results show that as droplet size increases, the variability of measurements decreases. This trend in interfacial tension was correlated with Bond number. However, to compare the results of different needle sizes, Worthington number was introduced to take into account the maximum volume allowed for a needle size **Berry2015**. Worthington number was defined as the ratio of a droplet volume to the maximum possible volume. Indeed, the maximum volume is another quantity that must be either defined or at least approximated. The necessity of approximation is a direct result of the unknown part of the droplet that stays on the needle after necking and droplet detachment. One way to define it is the theoretical maximum droplet that can be sustained **Garandet1994b**. The Worthington number can then be written as:

$$W_o = \frac{\Delta \rho g V_d}{\pi \gamma D_n} \tag{4.3}$$

where V_d is the drop volume, γ is the surface tension, and D_n is the needle tip diameter.

To investigate the drop volume effect on the measurements, a sequence of droplets were formed by using different needle sizes. This analysis essentially produces droplets with a range of Bond numbers. Using the setup and methodology described in Chapter 3, Figure 4.18 presents the change of surface tension of water when using three nozzles of varying sizes. Figure 4.19 shows the average values of surface tension using different needles grouped for three ranges of Worthington numbers. The largest needle size resulted in the most accurate fit compared to the known values of water surface tension.



Figure 4.18: The change of surface tension with apparent W_o number of a droplet suspended by three nozzles of different sizes.



Figure 4.19: The average values of surface tension when using different sized needles. The results are grouped using three different ranges of Worthington number.

In order to assess the stability of droplets as the drop size increases, Figure 4.20 shows the drop rotation angle against the apparent Worthington number. The variation appears to be minimal in the apparent W_o number of 0.4-0.7 for both needle sizes. Figure 4.21 shows the consecutive change in the rotation angle against Worthington number. This figure shows that minimum change occurs around $W_o = 0.5$. The values at small W_o results in considerable inconsistencies because the droplet shape tends to be spherical, leading to inaccurate results when using the pendant drop method. The droplet becomes unstable at large volumes, as indicated on the right side of the curves in Figure 4.21. At this stage, the droplet weight is high and starts to experience necking at the needle tip. The average values of surface tensions are also affected by the size of needle diameter as shown in Figure 4.22. The analysis ensured a good selection of the needle size and averaging range in the experimental results.



Figure 4.20: The rotation angle of images in the optimisation procedure to find the surface tension. The variance in the values gives an indication of the stability of the system at a particular Worthington number.



Figure 4.21: The consecutive change in the rotation angle against Worthington number. The minimum values occurs around $W_o = 0.5$.



Figure 4.22: Surface tension of water droplets for Worthington numbers larger than 0.5 using different sized needles. The figure also shows the average values of the presented data.

4.7.2 Surface tension of droplets with surface-loaded particles

The experimental results of pure and surface loaded droplets are summarised in Figure 4.23. The bars show the results for pure water and salt water at different mass concentrations. the mixture of salt and water has a dissimilar density than pure water. The amount of dissolved salt in water is called salinity, and is given by

$$S = \frac{m_s}{m_s + m_w} \tag{4.4}$$

where m_w (kg) is the pure water mass and m_s (kg) is the salt mass. For example, a salinity of 4% means that 4% of the total mass is salt and 96% is pure water. The salinity can be given in percentages, per mille, or even in parts per thousand.

Pure water surface tension dropped from 73 to 40 mN m⁻¹. This is a significant change of about 45% of the original value. Similar behaviour was observed in salt water when the surface was loaded with particles.



Figure 4.23: Surface tension of water droplets for Worthington numbers larger than 0.5 using different sized needles. The figure also shows the average values of the presented data.

4.7.3 Dynamic behaviour of surface tension

This section describes the behaviour of surface tension as the droplet volume is varied dynamically using the setup. Figure 4.24 shows the measured surface tension of a distilled water droplet. The volume of the droplet was increased in an interval of 10 seconds. This is indicated by the increase of the surface area from 28 to 46 mm². The surface tension of the droplet is almost constant with an average value of 72.6 mN m⁻¹.



Figure 4.24: Surface tension of a distilled water drop with varied drop volume.

The surface tension of a droplet with surface suspended particles on the surface changes with surface area. Figure 4.25 shows the surface tension of a droplet with surface suspended particles while increasing droplet volume at time intervals between 8 seconds to 16 seconds. During this period, the surface tension increases linearly with surface area. This increase implies that the concentration of particles on the surface is linked to the surface area of the droplet.



Figure 4.25: Surface tension change with drop volume increase. Two main behaviours of the droplet are indicated: fixed volume and increasing volume.

Following the previous results, further investigation of the dynamic behaviour was conducted. Figure 4.26 shows the surface tension of a pendant droplet with surface area fluctuations. Three points are indicated in figure: 1, 2, and 3. Point 1 is the start of droplet volume decrease where the surface tension starts to decrease. Point 2 is indicating the lowest volume and surface tension. Following point 2, the volume increases until point 3 where the surface tension seems to recover to a similar value of that at point 1.



Figure 4.26: The effect of volume fluctuation on the surface tension. Three different stages are marked with numbers 1, 2, and 3. The three different stages represent different stages of the droplet size and respective surface tension evolution.

Figure 4.27 shows the values of surface tension of the same droplet against Worthington number. The three points are also indicated in Figure 4.27 representing the same points that were discussed in Figure 4.26. Moving from point 1 to point 2, the surface tension decreases along with the surface area of the droplet. Point 2 shows a deflection as the surface tension starts to increase at a higher rate compared to to the region just before point 2. The values of surface tension did not follow the same path around point 2 and this indicates hysteresis in the surface behaviour. The particles on the surface may have reached a different arrangement as a result of the surface contraction.



Figure 4.27: The effect of volume fluctuation on the surface tension fluctuations at different Worthington number ranges.

4.8 Conclusions

An experiment was designed and executed successfully for droplet particle application, droplet evaporation, and surface tension measurements. The improved setup allowed for droplet temperature measurements using a fine thermocouple. In addition, the pendant drop tensiometry enhanced the results and enabled dynamic variation of droplet volume. The following results can be concluded:

- 1. The results of different smoke application times did not indicate a major change in the evaporation constant. This pointed towards the possibility of a maximum loading of particles on the surface of a droplet. Of course, this may depend on the available surface and particle characteristics.
- 2. The last stage of evaporation experienced lower evaporation constant compared to the major part of the droplet lifetime in all cases. While this is true for both pure and droplets with surface suspended particles, droplets with particles showed a higher evaporation constant. The enhanced evaporation suggests that

particles play a significant role when droplets are small or when they reach the final stages of evaporation.

- 3. Pendant drop tensiometry was used to measure the surface tension of particle loaded water droplets. Surface tension of loaded droplets was found to be different compared to pure water droplets. This behaviour can dramatically change the generation and interaction of water droplets in spray systems.
- 4. New results showed that the surface tension of droplets changed with droplet volume in contrast to pure droplets. The change in surface tension of droplets with surface suspended particles was studied dynamically by changing the droplet volume after particle loading.

Chapter 5

Droplet Evaporation and Effective Thermal Conductivity Modelling

This chapter aims to develop a numerical model to describe single droplet evaporation with suspended particles on its surface. The governing equations of mass and heat exchange are based on the evaporation of a pure water droplet. Furthermore, droplet evaporation parameters that were hypothesised to get affected by the addition of particles are discussed.

5.1 Introduction

Figure 5.1 illustrates how the thermal conductivity of particles and the area they occupy on the surface of a droplet can affect the evaporation process. The arrows in Figure 5.1 (a) point to a particle, a fluid medium, and a region which contains both particles and a fluid medium. The thermal conductivity of the particles and the fluid are donated by k_p and k_f , respectively. The effective thermal conductivity k_{eff} is thermal conductivity that can consider both the particles and the fluid properties when modelling heat transfer through such a medium.

Figure 5.1 (b) points to the surface area of the fluid A_f , the exposed area of particles A_p , and the addition of both contributions, which results in the overall effective evaporation area A.



Figure 5.1: Factors that may change the behaviour of droplet evaporation modelling when considering a droplet with surface suspended particles. Figure (a) points to a particle with thermal conductivity of k_p , a fluid with thermal conductivity of k_f , and a mixed region with an effective thermal conductivity of k_{eff} . Figure (b) points to the surface area contributions from both the particles and the fluid. The overall area is A, the exposed fluid area A_f , and the exposed area of particles A_p .

The formulation of a model that takes into consideration the physical changes mentioned above can be approached by the following steps:

- 1. Modelling of a droplet with suspended particles evaporation by considering a single droplet evaporation process.
- 2. Modelling of effective thermal conductivity.
- 3. Modelling the effective surface area available for evaporation by considering the wetting properties of the suspended particle.
- 4. Evaluating the effective conductivity and area models together with the droplet evaporation model.

Each step is an involved process where different parameters and physical situations must be considered. This chapter details the foundation of the first two steps. The droplet evaporation model was evaluated and compared with the experimental results. An extension of this chapter as future work is required to encompass the other two steps.

5.2 Theoretical foundation of single droplet evaporation

In contrast to the systems described in Chapter 2 and based on the morphological behaviours of droplets with volume-loaded particles, the evaporation of a droplet with surface particles may not necessarily pass through many stages. The main difference is that surface-loaded droplets have a considerably lower chance of forming a shell at the first stages because of the relatively small number of particles compared to the volume-loaded droplets. Therefore, the evaporation of a pure water droplet can be used to model the current system. Furthermore, considering the possibility of a shell formation as the droplet evaporates, two models can be assessed:

- 1. Shell formation: the evaporation process can be divided into two stages, the first stages where the evaporation progress until a second stage when the clusters of particles from a solid shell. This was the observation in the literature related to volume particle loading and when using solute materials. In the experiments of this current research, shell formation was not observed.
- 2. No shell formation model: the amount of particle loading does not lead to this phenomenon. This was the main observation in the experiments of this current work.

The analysis given in this chapter follows the assumption of no shell formation. This is justified by the experimental observations detailed in Section 2.5.4, Chapter 3, and in Chapter 4.

For a one dimensional model of the temperature inside a droplet, the heat transfer governing equation can be written as:

$$\rho_d c_{p_r d} \frac{\partial T}{\partial t} = \frac{k_d}{r^2} \frac{\partial}{\partial r} \left(r^2 \frac{\partial T}{\partial r} \right)$$
(5.1)

Droplet properties such as density ρ_d (kg/m³), thermal conductivity k_d (W/m · K), and specific heat capacity $c_{p,d}$ (J/kg · K) are determined using the water properties at droplet reference temperature. Temperature distribution within the droplet T (K), is calculated by solving equation 5.1 under the following boundary conditions:

$$\frac{\partial T}{\partial r} = 0 \quad \text{at} \quad r = 0$$

$$h\left(T_g - T_d\right) = k_d \frac{\partial T}{\partial r} + h_{fg} \frac{\dot{m}_v}{A_d} \quad \text{at} \quad r = r_d$$
(5.2)

where r_d (m) and A_d (m²) are the outer radius and surface area of the droplet, h_{fg} (J/kg) is the specific heat of evaporation, T_g (K) and T_d (K) are hot air and droplet surface temperatures, and \dot{m}_v (kg/s) is the mass transfer rate due to evaporation. Convective heat transfer coefficient h is calculated using the Ranz and Marshall correlation for Nusselt number [5]:

$$Nu = \frac{2r_d h}{k_g} = 2 + 0.6 Re^{1/2} Pr^{1/3}$$
(5.3)

where Re and Pr are the Reynolds and Prandtl numbers, respectively, and k_g (W/m · K) is thermal conductivity of hot air. Evaporation rate, \dot{m}_v (kg/s), is related to surface radius, r_d (m), which can be evaluated using:

$$\dot{m}_{\rm v} = 4\pi r_s \left(\frac{k_g}{c_{\rm p,g}}\right) \ln\left(1 + B_{\rm M}\right)$$

$$B_{\rm M} = \frac{Y_{w,s} - Y_{w,\infty}}{1 - Y_{w,s}}$$
(5.4)

where $Y_{w,s}$ and $Y_{w,\infty}$ are water vapour mass fraction at the droplet surface and at the free stream, respectively. Here B_M is the mass transfer number and it represents the driving force for evaporation. Droplet radius changes due to evaporation is calculated using

$$\frac{dr_d}{dt} = -\frac{1}{4\rho_d \pi r_d^2} \dot{m}_v \tag{5.5}$$

5.3 Explicit method discretisation

The explicit discretisation depends on space derivatives evaluated in a previous time step. This results in overall equations that depend only on the results of the previous steps. The governing equation is applied at each space step and is solved in turn, starting from the initial conditions. In the following equations, space discretisation is donated by the subscript i, and time discretisation is donated by the superscript n. For example, the term T_i^n is the temperature at space step i and current time step n, while T_{i+1}^{n-1} is the temperature at the next space step i+1 and the previous time step n-1. Furthermore, the space step size is donated by Δr , and the time step size is donated by Δt . Starting with governing one-dimensional equation 5.1, the derivative with respect to the radius on the right side of the equation can be rewritten as follows:

$$\rho_d c_{p,d} \frac{\partial T}{\partial t} = \frac{k_d}{r^2} \frac{\partial}{\partial r} \left(r^2 \frac{\partial T}{\partial r} \right)$$

$$\rho_d c_{p,d} \frac{\partial T}{\partial t} = k_d \left(\frac{\partial^2 T}{\partial r^2} + \frac{2}{r} \frac{\partial T}{\partial r} \right)$$
(5.6)

Using the laws of first derivative discretisation, the derivatives with respect to time and radius can be stated as:

$$\frac{T_i^n - T_i^{n-1}}{\Delta t} = \frac{k_d}{\rho_d c_{p,d}} \left(\frac{T_{i-1}^{n-1} - 2T_i^{n-1} + T_{i+1}^{n-1}}{\Delta r^2} + \frac{2}{r} \times \frac{T_{i+1}^{n-1} - T_i^{n-1}}{\Delta r} \right)
T_i^n = T_i^{n-1} + \frac{\Delta t k_d}{\rho_d c_{p,d}} \left(\frac{T_{i-1}^{n-1} - 2T_i^{n-1} + T_{i+1}^{n-1}}{\Delta r^2} + \frac{2}{r} \times \frac{T_{i+1}^{n-1} - T_i^{n-1}}{\Delta r} \right)$$
(5.7)

where the backward in time discrete form was used for the time derivative, central in space form for the second order derivative with respect to radius, and forward in space for the first order derivative with respect to radius. All the time steps in the derivatives with respect to radius were in the previous time step, so the explicit discretisation remains applicable.

5.3.1 Boundary conditions

The first boundary condition is concerned with the symmetry of the temperature profile at the centre of the droplet. This is a direct result of the assumption that the droplet is spherical and that the heat transfer and environment condition are homogeneous. This boundary condition is stated by:

$$\frac{\partial T}{\partial r} = 0 \tag{5.8}$$

Since this boundary is at the start of the domain, which is the droplet centre, the mirror principle which deals with symmetry boundary conditions can be utilised as:

$$\frac{T_{i-1}^n - T_i^n}{\Delta r} = 0 \to \frac{T_{i+1}^n - T_i^n}{\Delta r} = 0 \to T_i^n = T_{i+1}^n$$
(5.9)

The mirror principle ensures zero gradient at the boundary. The second boundary is at the surface of the droplet which is more involved since it has to include the fact that some liquid becomes vapour. The second boundary condition is:

$$h\left(T_g - T_d\right) = k_d \frac{\partial T}{\partial r} + h_{fg} \frac{\dot{m}_v}{A_d}$$
(5.10)

The left part of the equation above represent the thermal energy need to raise the droplet temperature T_d to the gas temperature T_g which is the temperature of the droplet's surroundings. The first term on the right side is conducted heat towards the centre of the droplet, and the second term is the energy lost due to evaporation. The derivative with respect to radius can be approximated by the backward difference. This boundary can then be rearranged as following:

$$\frac{k_g}{r} \left(T_g - T_i^n \right) = k_d \frac{T_i^n - T_{i-1}^n}{\Delta r} + h_{fg} \frac{\dot{m}_v}{A_d}
\frac{k_g}{r} T_g - \frac{k_g}{r} T_i^n = \frac{k_d}{\Delta r} T_i^n - \frac{k_d}{\Delta r} T_{i-1}^n + h_{fg} \frac{\dot{m}_v}{A_d}
- \frac{k_g}{r} T_i^n - \frac{k_d}{\Delta r} T_i^n = \frac{k_d}{\Delta r} T_i^n - \frac{k_d}{\Delta r} T_{i-1}^n + h_{fg} \frac{\dot{m}_v}{A_d} - \frac{k_g}{r} T_g
T_i^n \left(-\frac{k_g}{r} - \frac{k_d}{\Delta r} \right) = -\frac{k_d}{\Delta r} T_{i-1}^n + h_{fg} \frac{\dot{m}_v}{A_d} - \frac{k_g}{r} T_g$$
(5.11)

Finally it can be written as:

$$T_i^n = \left(-\frac{k_d}{\Delta r}T_{i-1}^n + h_{fg}\frac{\dot{m}_v}{A_d} - \frac{k_g}{r}T_g\right) / \left(-\frac{k_g}{r} - \frac{k_d}{\Delta r}\right)$$
(5.12)

The equation above depends on the temperature that is evaluated at a previous space step during the same time step.

5.4 Implicit method discretisation

The implicit method forms a system of equations for all points in the domain. These equations are dependent on each other and can only be evaluated if they are solved at once. Usually, these equations are put into a matrix form so that matrix operations can be applied to evaluate the results. To write the equations for the implicit method, the space derivatives are no longer evaluated at a previous time step, but rather at the current time step.

For the space domain i > 0 and $i < r_d$, Equation 5.1 can be discretised as follows:

$$\frac{T_i^n - T_i^{n-1}}{\Delta t} = \frac{k_d}{\rho_d c_{p,d}} \left(\frac{T_{i-1}^n - 2T_i^n + T_{i+1}^n}{\Delta r^2} + \frac{2}{i\Delta r} \times \frac{T_{i+1}^n - T_i^n}{\Delta r} \right)$$
(5.13)

Note that only a single term evaluated during the past time step, which appears on in time derivative approximation. On the other hand, all other terms are in the current time step which have not been evaluated yet. This shows that the implicit method requires a set of equations for the whole domain which can be solved simultaneously. In order to use matrix operations, Equation 5.13 can be rearranged as follows:

$$T_{i}^{n} - \frac{\Delta t \times k_{d}}{\rho_{d}c_{p,d}} \left(\frac{T_{i-1}^{n} - 2T_{i}^{n} + T_{i+1}^{n}}{\Delta r^{2}} + \frac{2}{i} \times \frac{T_{i+1}^{n} - T_{i}^{n}}{\Delta r^{2}} \right) = T_{i}^{n-1}$$

$$T_{i}^{n} - \frac{\Delta t \times k_{d}}{\rho_{d}c_{p,d}\Delta r^{2}} \left(T_{i-1}^{n} - 2T_{i}^{n} + T_{i+1}^{n} + \frac{2}{i} \times T_{i+1}^{n} - \frac{2}{i} \times T_{i}^{n} \right) = T_{i}^{n-1}$$

$$T_{i}^{n} - \frac{\Delta t \times k_{d}}{\rho_{d}c_{p,d}\Delta r^{2}} \left(T_{i-1}^{n} - \left(2 + \frac{2}{i} \right) \times T_{i}^{n} + \left(1 + \frac{2}{i} \right) \times T_{i+1}^{n} \right) = T_{i}^{n-1}$$
(5.14)

The final equation which can be arranged into a diagonal matrix is:

$$-\frac{\Delta t \times k_d}{\rho_d c_{p,d} \Delta r^2} \left(T_{i-1}^n - \left(2 + \frac{2}{i} + \frac{\rho_d c_{p,d} \Delta r^2}{\Delta t \times k_d} \right) \times T_i^n + \left(1 + \frac{2}{i} \right) \times T_{i+1}^n \right) = T_i^{n-1}$$

$$\tag{5.15}$$

5.4.1 Boundary conditions

Since equation 5.15 is second order in space, two boundary conditions are required. For i = 0, the boundary condition at the centre of the droplet is symmetry, in other words the gradient of temperature profile is zero:

$$\frac{\partial \mathbf{T}}{\partial r} = 0$$

$$\frac{T_i^n - T_{i-1}^n}{\Delta r} = 0$$
(5.16)

Applying this boundary condition in equation 5.15:

$$-\frac{\Delta t \times k_d}{\rho_d c_{p,d} \Delta r^2} \left(-\left(1 + \frac{2}{i} + \frac{\rho_d c_{p,d} \Delta r^2}{\Delta t \times k_d}\right) \times T_i^n + \left(1 + \frac{2}{i}\right) \times T_{i+1}^n \right) = T_i^{n-1} \quad (5.17)$$

The second boundary condition is at the droplet surface where mass and heat is transferred. If the temperature in the environment is higher than the droplet temperature, which is the case in a fire extinguishment scenario, heat is transferred from the environment to the surface of the droplet. In addition, evaporation occurs at the surface which leads to a loss of latent heat. Heat condition in the liquid side also occurs at the surface. The boundary condition can be written as:

$$T_{i+1}^n = \left(-\frac{k_d}{\Delta r}T_i^n + h_{fg}\frac{\dot{m}_v}{A_d} - \frac{k_g}{r}T_g\right) / \left(-\frac{k_g}{r} - \frac{k_d}{\Delta r}\right)$$
(5.18)

Equation 5.18 has to be substituted in 5.15. A substation is made below to simply the mathematical rearrangements. Let

$$\delta = \left(h_{fg}\frac{\dot{m}_v}{A_d} - \frac{k_g}{r}T_g\right) / \left(-\frac{k_g}{r} - \frac{k_d}{\Delta r}\right)$$

$$\vartheta = -\frac{k_d}{\Delta r} / \left(-\frac{k_g}{r} - \frac{k_d}{\Delta r}\right)$$
(5.19)

Equation 5.18 can then be written as:

$$T_{i+1}^n = \vartheta \times T_i^n + \delta \tag{5.20}$$

Evaluating equation 5.20 in equation 5.15 results in:

$$-\frac{\Delta t \times k_d}{\rho_d c_{p,d} \Delta r^2} \left(T_{i-1}^n - \left(2 + \frac{2}{i} + \frac{\rho_d c_{p,d} \Delta r^2}{\Delta t \times k_d} - \left(1 + \frac{2}{i} \right) \times \vartheta \right) \times T_i^n + \left(1 + \frac{2}{i} \right) \times \delta \right) = T_i^{n-1}$$
(5.21)

$$-\frac{\Delta t \times k_d}{\rho_d c_{p,d} \Delta r^2} \left(T_{i-1}^n - \left(2 + \frac{2}{i} + \frac{\rho_d c_{p,d} \Delta r^2}{\Delta t \times k_d} \right) \times T_i^n + \left(1 + \frac{2}{i} \right) \times \left(\vartheta \times T_i^n + \delta \right) \right) = T_i^{n-1} \quad (5.22)$$

Boundary condition 1

$$\frac{\partial T}{\partial r} = 0 \tag{5.23}$$

Applying the mirror principle on the symmetry line:

$$\frac{T_{i-1}^n - T_i^n}{\Delta r} = 0 \to \frac{T_{i+1}^n - T_i^n}{\Delta r} = 0 \to T_i^n = T_{i+1}^n$$
(5.24)

The second boundary condition is concerned with the energy balance at the surface droplet. Similar to equation 5.10, the boundary condition at $r = r_s$ can be stated as:

$$h\left(T_g - T_d\right) = k_d \frac{\partial T}{\partial r} + h_{fg} \frac{\dot{m}_v}{A_d}$$
(5.25)

The equation above can be discretised and rearranged as follows:

$$\frac{k_g}{r} \left(T_g - T_i^n \right) = k_d \frac{T_i^n - T_{i-1}^n}{\Delta r} + h_{fg} \frac{\dot{m}_v}{A_d}$$

$$\frac{k_g}{r} T_g - \frac{k_g}{r} T_i^n = \frac{k_d}{\Delta r} T_i^n - \frac{k_d}{\Delta r} T_{i-1}^n + h_{fg} \frac{\dot{m}_v}{A_d}$$

$$-\frac{k_g}{r} T_i^n - \frac{k_d}{\Delta r} T_i^n = \frac{k_d}{\Delta r} T_i^n - \frac{k_d}{\Delta r} T_{i-1}^n + h_{fg} \frac{\dot{m}_v}{A_d} - \frac{k_g}{r} T_g$$

$$T_i^n \left(-\frac{k_g}{r} - \frac{k_d}{\Delta r} \right) = -\frac{k_d}{\Delta r} T_{i-1}^n + h_{fg} \frac{\dot{m}_v}{A_d} - \frac{k_g}{r} T_g$$
(5.26)

The second boundary condition in the final form is

$$T_i^n = \left(-\frac{k_d}{\Delta r}T_{i-1}^n + h_{fg}\frac{\dot{m}_v}{A_d} - \frac{k_g}{r}T_g\right) / \left(-\frac{k_g}{r} - \frac{k_d}{\Delta r}\right)$$
(5.27)

The equation above requires the temperature at a previous space step during the same timestep. But since all the temperature values are all evaluated simultaneously, this boundary condition equation also needs to be included in the implicit matrix.

MATLAB was used to evaluate the model equations with the ability to change initial conditions. Figure 5.2 is a flow illustration of the script that was constructed and used in the evaluation of the results. The code starts with inputs of the initial droplet temperature, relative humidity, environment temperature, and droplet and air properties. This is followed by setting the spatial and temporal discretisation steps. The script then enters a loop to evaluate the properties, prepare the matrix of governing equations, set the boundary conditions, solve the matrix, and update the evaporation parameters. NIST Reference Fluid Thermodynamic and Transport Properties Database (REFPROP) Version 10 LEMMON-RP10 was used to evaluate the thermodynamic properties of air and water. The evaporation parameters include the mass loss and the reduction in droplet radius. The code checks whether the droplet radius has reached a very small value or when the mass of the droplet becomes a negative value. The loop is restarted again if the droplet has not evaporated completely. On the other hand, the code terminates if the radius reaches a threshold value. The results were compared to experimental results, as detailed below in the next section.



Figure 5.2: Flow of script which was written in MATLAB.

5.5 Droplet evaporation under room and elevated temperature conditions

The droplet evaporation model was validated using experimental results. The experimental setup was detailed in Chapter 4. Figure 5.3 presents the evolution of droplet diameter and temperature at room temperature conditions. The initial droplet diameter was $D_o = 1.2 \ mm$ and the environment temperature and relative humidity were 22 °C and 35%, respectively. Figure 5.3 (a) indicates that the evaporation behaviour follows the d-squared law and that the current model fits the experimental data. The experimental curve deviates form the model around $t/D_o^2 = 350 \ s/mm^2$. This deviation may be a result of the long time taken for evaporation and the change of environmental parameters.

Figure 5.3 (b) is a comparison between the recorded and modelled droplet temperature. The initial experimental droplet temperature is 16°C which is lower than the environment temperature. The explanation is that the droplet is continuously evaporating, and this is the steady state temperature. If the initial temperature of the droplet was set to be the same as that of the room temperature, the drop in temperature could be captured as shown by the model curve in Figure 5.3 (b). Starting from 350 s/mm^2 , the experimental temperature started to increase. This was probably due to the change in the environmental condition as reflected in the diameter progression as well. The significant variations in the temperature at the end of the experimental curve were due to the droplet becoming increasingly small so that it detaches from the thermocouple. At this stage, the thermocouple was reading the temperature of the surroundings.

In addition to the room temperature comparisons that were shown above, the droplet evaporation model was tested against the experimental results of droplet evaporation under heated conditions. Figure 5.4 shows the evolution of droplet diameter and temperature at elevated surrounding temperatures of $T_g = 230$ °C. The initial droplet diameter was $D_o = 1.2 mm$, and the environment relative humidity was 30% measured at room temperature of 23 °C. Figure 5.4(a) indicates that the evaporation behaviour follows the d-squared law and that the current model fits the experimental data. The model predicted the behaviour of droplet diameter evolution at elevated temperatures. However, the model steady state temperature was lower compared to experimental results by 5 °C approximately. This is possibly due to the additional heating from the thermocouple wire [213]. Furthermore, The model captured the behaviour of droplet temperature increase at the beginning of the heating process as shown in Figure 5.4 (b). In the following section, an examination of the effect of particles on thermal conductivity is presented. First, the most fundamental parallel and series models are presented. This is followed by a detailed derivation of one-dimensional models for the current investigated case. Finally, a derivation of Maxwell's model based on heat transfer theory is presented, along with a discussion about the assumptions and applicability.



(b) Droplet temperature

Figure 5.3: Evolution of a droplet diameter and temperature with initial diameter $D_o = 1.2mm$ at room conditions of 22 °C environment temperature and 35% relative humidity.





Figure 5.4: Evolution of droplet diameter and temperature at elevated surrounding temperature. The initial droplet diameter was $D_o = 1.2mm$ at a relative humidity of 30% measured at room temperature of 23 °C. The initial droplet temperature was $T_d(t=0) = 13$ °C. The droplet surrounding temperature was $T_g = 230$ °C.

5.6 Effect of suspended particles on thermal conductivity

Following the modelling of evaporating droplets, the effect of particles on thermal conductivity is considered. The addition of solid particles into liquids is known to affect the properties of the liquids. Thermal conductivity is one that can increase, by a significant amount, despite the low fraction of particles when mixed with liquids. Studies have shown a dependence on particle size, shape, and concentration. The level of variability in the published results presents a difficulty in understanding the mechanism of heat transfer. A benchmark study conducted to study stable dispersions thermal conductivity using a wide variety of methods concluded that the results were comparable within 10% of the sample average [214].

One of the possible reasons that has been identified is the inability to control the dispersion of particles. Aggregation is expected to play a role in variabilities and can explain contradicting results [215]. Clusters of particles have been shown to result in efficient heat transport and therefore higher conductivity, experimental investigations concluded that an increase of aggregate size at the same particle fraction is correlated with higher effective conductivity.

Similar to the subject of dispersed particles in fluids is the mixture of solid composites. An important difference appears in the conductivity performance of solid composites. The thermal conductivity is reduced below that of the continuous medium when smaller particles are added in some cases [216]. This is explained by the interfacial resistance in between the filler and the base medium. Interfacial resistance was found to be inversely proportional to the particle size. Hence, introducing smaller particles into solids leads to higher interfacial resistance and lower effective thermal conductivity [217].

The most fundamental models for the effective thermal conductivity are the series and parallel resistance models. Considering a control volume in which the left side is a heat source, and the right side is a heat sink, the series model as shown in Figure 5.5 (a) assumes that the transfer of heat occurs in one path through the first medium and then the other. While the parallel model presented in Figure 5.5 (b) assumes that part of the heat transfers through the particles and the other part through the fluid at the same time.

The two models can be derived through the thermal resistance analysis. Starting with the series model, let the particles be represented by a cross sectional area A_p (m²), a length of L_p (m) and a conductivity k_p (W/m · K). In a similar way, let A_f



Figure 5.5: Series and parallel models representing two extremes when analysing the effective thermal conductivity as one chain. The particles are illustrated by the solid circles while the surrounding blue area is the fluid. The most fundamental series and parallel model assume no contact resistance between particles.

 (m^2) , L_f (m) and k_f (W/m · K) be the cross sectional area, length and conductivity of the fluid respectively. The total thermal resistance of two mediums in series can be written as:

$$R_{eff} = R_p + R_f \tag{5.28}$$

If we assume a one-dimensional heat transfer through a plane, the effective resistance becomes

$$\frac{L}{k_{eff}A} = \frac{L_p}{k_p A_p} + \frac{L_f}{k_f A_f}$$
(5.29)

Given that this is a series model, the area is the same through out $A = A_p = A_f$. By defining the particles volume ratio to overall volume $\phi = \frac{V_p}{V} = \frac{AL_p}{AL} = \frac{L_p}{L}$, Equation 5.29 then becomes:

$$\frac{1}{k_{eff}} = \frac{\phi}{k_p} + \frac{(1-\phi)}{k_f}$$
(5.30)

Rearranging to find the effective thermal conductivity

$$k_{eff} = \frac{k_p k_f}{\phi k_f + (1 - \phi) k_p} \tag{5.31}$$

The parallel model differs in that that the cross sectional area of particles is not the same as the overall area. However the length for both mediums is the same $L = L_p = L_f$. The effective resistance is

$$\frac{1}{R_{eff}} = \frac{1}{R_p} + \frac{1}{R_f}$$
(5.32)

Similar to the assumption of one-dimensional plane heat conduction in the series model, the resistance terms can be substituted in Equation 5.32:

$$\frac{k_{eff}A}{L} = \frac{k_pA_p}{L_p} + \frac{k_fA_f}{L_f}$$
(5.33)

Using the volume concentration of particles $\phi = \frac{V_p}{V} = \frac{A_p L}{AL} = \frac{A_p}{A}$, Equation 5.32 can be simplified as

$$k_{eff} = \phi k_p + (1 - \phi) k_f \tag{5.34}$$

Although the parallel model has been shown to describe the behaviour of tube fibres when aligned parallel to the heat transfer direction, both series and parallel models do not apply strictly to most fluid suspension mixtures.

The basic series and parallel models represent the limits for a possible binary mixture. The parallel model allows the most preferable path for maximum heat transfer. On the other hand, the equation of effective thermal conductivity in the series configuration shows that the results will be either that of the medium with the minimum conductivity or even lower than that of both mediums. In other words, the effective thermal conductivity in the series model cannot be more than that of any constituent.

The classical model for the effective conductivity of dispersed particles is Maxwell's model, a theory that was originally developed to evaluate the conduction in heterogeneous media [201], the following equation is the result of Maxwell analysis:

$$k_{eff} = k_f \frac{2k_f + k_p + 2\phi(k_p - k_f)}{2k_f + k_p - \phi(k_p - k_f)}$$
(5.35)

The model is based on well dispersed spherical particles in a larger sphere of another medium. Equation 5.35 shows that k_{eff} tends to k_f as the volume fraction $\phi \to 0$, while the k_{eff} will be that of the particles k_p as $\phi \to 1$. But the model assumes low volume particle fraction, and once the concentration is high enough [214], the temperature field around one particle is affected by the nearby particles.





(a) Random distribution of particles in a medium

(b) Particle clusters in a medium

Figure 5.6: Arrangements of particles in a medium can take different shapes spanning from random distribution to clusters. Different arrangements can influence the type of heat transfer system and thermal resistances between particles.

The original derivation of Equation 5.35 considered two concentric spheres, an outer sphere with conductivity k_f and an inner sphere with conductivity k_p . Now, assume the arrangement of the two phases is reversed, Equation 5.35 can be modified to reflect the effective conductivity of the new system. By swapping k_f and k_p and replacing ϕ with $(1 - \phi)$, 5.35 becomes:

$$k_{eff} = k_p \frac{2k_p + k_f - 2(1 - \phi)(k_p - k_f)}{2k_p + k_f + (1 - \phi)(k_p - k_f)}$$
(5.36)

Equations 5.35 and 5.36 represents two limits on the conductivity of binary mixtures of particles and liquids at the same volume fraction. Figure 5.6 (a) illustrates the lower limit in Equation 5.35 which shows dispersed particles in the surrounding medium. Equation 5.36 physically represent a configuration shown in Figure 5.6 (b) where the particles are arranged in chains [217] or solid network with liquid pockets [215]. It is worth noting that these two bounds are in fact the same as Hashin and Shtrikman model which cannot be improved without additional information on the statistical variation of the particles [218].

Several models have been proposed to include the effect of the interfacial layer surrounding the particles [219], [220]. The approach usually takes into account the thermal resistance of a thin layer on the particles. This layer has a thermal conductivity higher than that of liquid but lower than the particle conductivity. The models are claimed to result in improved predictions compared to Maxwell model. However, the effect due to this ordered layer is negligible in most suspensions as concluded in [217]. This is of course different for solid composites where the effective conductivity is strongly dependent on the size of fillers and the interfacial layer as mentioned above.

5.7 Thermal resistance models

The development of two models are described in the section. Figure 5.7 shows the system in consideration, a droplet with suspended particles on its surface. In the extended view of Figure 5.7, the one dimensional domain contains particles on the left surrounded by a fluid medium, this is then followed by a fluid region. A heat source is assumed at the left edge, while a heat sink is assumed at the right side. The task is to find the effective thermal conductivity for the system in consideration.



Figure 5.7: Illustration of the one dimensional assumption when modelling the thermal conductivity of the particles and fluid system. The heat source is assumed on the side of the particles as the particles are suspended on the surface.

To study the case presented in Figure 5.7 using thermal resistance analysis, two possible configurations are illustrated in Figure 5.8. The arrangements of resistances depend on the mediums, thermal properties, physical arrangement, and the joint points of different mediums. Diagrams of the two possible arrangements in Figure 5.8 show the physical positions of the particles, fluid, contact lines, and an indication of areas. Furthermore, thermal resistance circuits are shown along with the arrangement



Figure 5.8: Two variants of one-dimensional effective conductivity models using thermal resistance concept. The particles are represented by the solid black circles while the fluid medium is represented by the blue regions. The white circles represent that the area is filled by either the particles or the fluid.

diagrams. The circuits indicate how different sections of the system are connected. Individual resistances in the circuits can be connected in series or in parallel with other resistances.

Formulating the effective thermal resistance required the definition of area ratio of particles A_p , to full area A exposed to the heat source which can be written as:

$$\frac{A_p}{A} = \phi \tag{5.37}$$

$$\frac{A_f}{A} = 1 - \phi$$

The later equation is the area ratio of the fluid A_f to the total exposed area. Figure 5.9 is a schematic of the model which illustrates the definition of the area ratios. In addition, the thickness of the particle layer is also shown on Figure 5.9.



Figure 5.9: A schematic of area and layer thickness parameters used in the effective thermal conductivity model.

5.7.1 Model 1

Considering the model shown in Figure 5.8, the network is constructed where the resistances are divided into two parallel paths. The bottom path is constructed out of two resistances in series, a particle layer resistance donated by p followed by a fluid resistance donated by f_1 that spans to the heat sink. The top path represents a fluid section to the heat sink on the right and is donated by the subscript f_2 . The top path has a length of L and an area of A_f . On the other hand, the bottom path has an area A_p , the particle layer and fluid series resistances have a length of ψL and $(1 - \psi)L$, respectively. The terms of different resistances are listed below:

$$R_{p} = \frac{\psi L}{k_{p}A_{p}}$$

$$R_{f1} = \frac{(1-\psi)L}{k_{f}A_{p}}$$

$$R_{f2} = \frac{L}{k_{f}A_{f}}$$

$$R_{eff} = (\frac{1}{R_{p}+R_{f_{1}}} + \frac{1}{R_{f_{2}}})^{-1}$$
(5.38)

The problem is solved in two steps, starting with the bottom path. The two resistances in series can be added as follows:

$$R_{p+f_1} = R_p + R_{f_1} \tag{5.39}$$

Later, the two parallel paths R_{p+f_1} and R_{f_2} and can be added using parallel resistance rule:

$$R_{eff} = \left(\frac{1}{R_{p+f_1}} + \frac{1}{R_{f2}}\right)^{-1} \tag{5.40}$$

Substituting the terms for resistances in Equation 5.39 as stated in Figure 5.8 results in

$$\frac{L}{k_{p+f_1}A_p} = \frac{\psi L}{k_p A_p} + \frac{(1-\psi)L}{k_f A_p} = \frac{\psi L k_f A_p + (1-\psi)L k_p A_p}{k_p A_p k_f A_p}$$
(5.41)

The series path effective thermal conductivity can then be stated as:

$$\frac{1}{k_{p+f_1}} = \frac{\psi k_f + (1-\psi)k_p}{k_p k_f} \tag{5.42}$$

Now, the two parallel paths can be solved as follows:

$$\frac{1}{R_{eff}} = \frac{1}{R_{f2}} + \frac{1}{R_{p+f_1}}$$

$$\frac{k_{eff}A}{L} = \frac{K_fA_f}{L} + \frac{K_pK_f}{\psi k_f + (1-\psi)k_p} \cdot \frac{A_p}{L}$$

$$k_{eff} = k_f(1-\phi) + \frac{\phi k_pk_f}{\psi k_f + (1-\psi)k_p}$$

$$\frac{k_{eff}}{k_f} = 1 + \frac{\phi \psi(\frac{k_p}{k_f} - 1)}{\psi + (1-\psi)\frac{k_p}{k_f}}$$
(5.43)

Finally, using the ratio of particle to fluid thermal conductivity $r = \frac{k_p}{k_f}$, it can be presented in the following form:

$$\frac{k_{eff}}{k_f} = 1 + \frac{\phi\psi(r-1)}{\psi + (1-\psi)r}$$
(5.44)

In the limit of $\psi = 0$, the effective conductivity is the same as that of the fluid, in other words, the particle layer vanishes. And in the limit of $\psi = 1$, the equation for a continuous particle chain is recovered. Furthermore, in the limit of high values of r, the equation becomes insensitive to the ratio of thermal conductivity values r, and Equation 5.44 is reduced to:

$$\frac{k_{eff}}{k_f} = 1 + \frac{\psi\phi}{(1-\psi)}$$
(5.45)

Figure 5.10 shows the change of k_{eff}/k_f as a function of ψ for different values of r, the area ratio ϕ is set to 0.15. As r becomes high, the curves manifest Equation 5.45, and the effective conductivity becomes insensitive to r. Furthermore, the increase of effective conductivity is highly predominant when the particle layer becomes comparable to the fluid only section, as Figure 5.10 shows a sharp increase for $\psi > 0.6$. The trend implies that the effect of smoke particles on the surface might be significant for smaller droplets. And that a droplet might experience an increase in overall conductivity as it evaporates.



Figure 5.10: The effect of particle layer thickness on the effective thermal conductivity for a range of particle to fluid thermal conductivity ratios. The area ratio $\phi = 0.15$ was used to present model 1 results.

5.7.2 Model 2

In this model, the heat source is connected to two parallel resistances first. The top part of these two resistances is the fluid section, donated by f_1 with area A_f and length ψL . The bottom part of two resistances in parallel is the particles layer resistance, donated by p with a an area of A_p and a length of ψL . These two parallel resistances are followed by a fluid resistance, donated by f_2 in series with the first section. The later resistance has an area same to the overall area A and a length of $(1 - \psi)L$. These resistances are stated below:
$$R_{p} = \frac{\psi L}{k_{p}A_{p}}$$

$$R_{f1} = \frac{\psi L}{k_{f}A_{f}}$$

$$R_{f2} = \frac{(1-\psi)L}{k_{f}A}$$
(5.46)

The overall effective resistance is:

$$R_{eff} = \left(\frac{1}{R_p} + \frac{1}{R_{f1}}\right)^{-1} + R_{f2}$$
(5.47)

This equation can be solved into two steps, starting with the first section of the resistances in parallel R_{p+f1}

$$R_{eff} = R_{p+f1} + R_{f2}$$

$$R_{p+f1} = \left(\frac{1}{R_p} + \frac{1}{R_{f1}}\right)^{-1}$$
(5.48)

substituting the terms of the resistances R_p and R_{f1} :

$$R_{p+f1} = \frac{k_p A_p}{\psi L} + \frac{k_f A_f}{\psi L}$$

$$\frac{k_{p+f1} A}{\psi L} = \frac{k_p A_p}{\psi L} + \frac{k_f A_f}{\psi L}$$
(5.49)

The equation of the combined resistance R_{p+f1} can then be written as:

$$k_{p+f1} = \phi k_p + (1 - \phi) k_f \tag{5.50}$$

Adding resistance R_{p+f1} and R_{f2} using the series rule and substituting the terms for each resistance:

$$R_{eff} = R_{p+f1} + R_{f2}$$

$$\frac{L}{k_{eff} A} = \frac{\psi L}{k_{p+f1}A} + \frac{(1-\psi)L}{k_f A}$$

$$\frac{1}{k_{eff}} = \frac{\psi}{k_{p+f1}} + \frac{(1-\psi)}{k_f} = \frac{\psi k_f + (1-\psi)k_{p+f1}}{k_{p+f1}k_f}$$

$$k_{eff} = \frac{k_{p+f1}k_f}{\psi k_f + (1-\psi)k_{p+f1}}$$

$$\frac{k_{eff}}{k_f} = \frac{\phi k_p + (1-\phi)k_f}{\psi k_f + (1-\psi)(\phi k_p + (1-\phi)k_f)}$$
(5.51)

Finally, the effective thermal conductivity based on model 2 can be written as:

$$\frac{k_{eff}}{k_f} = \frac{\phi r + (1 - \phi)}{\psi + (1 - \psi)(\phi r + (1 - \phi))}$$
(5.52)

In the limit of $\psi = 0$, the effective conductivity is the same as that of the fluid, in other words, the particle layer vanishes. And in the limit of $\psi = 1$, the equation for a continuous particle chain is recovered. Theses two results are the same for model 1 Equation 5.44. Both model 1 and 2 lead to the same equation when there is very small particle layer or when a continuous particle chain is formed between the heat source and heat sink. In addition, in the limit of high values of r, the equation becomes independent of r. Therefore, Equation 5.52 is reduced to:

$$\frac{k_{eff}}{k_f} = \frac{1}{1 - \psi}$$
 (5.53)

Figure 5.11 shows the change of k_{eff}/k_f as a function of ψ for different values of particle fluid thermal conductivity ratio r and an area ratio ϕ of 0.15. The curves tend to Equation 5.53 as r increases, and the effective conductivity becomes insensitive to r. In this model, the effective thermal conductivity change is faster in comparison to model 1.



Figure 5.11: The effect of particle layer thickness on the effective thermal conductivity for a range of particle to fluid thermal conductivity ratios. The area ratio $\phi = 0.15$ was used to present model 2 results.

Figure 5.12 is a comparison of model 1 and 2 effective thermal conductivity as a function of ψ at $\phi = 0.15$ and r = 10. Model 1 presents a lower limit of the current analysis while model 2 forms an upper limit of the effective thermal conductivity. This behaviour is similar to the most fundamental serial and parallel configuration discussed earlier. The analysis above assumes that the temperature gradients in other dimensions are negligible, and that one dimensional heat transfer is applicable. This is valid if the particles are distributed evenly in the radial axis and that the variation in the thermal conductivity of particles is not significant.



Figure 5.12: A comparison of model 1 and 2 results on the effect of particle layer thickness on the effective thermal conductivity for r = 10. The area ratio $\phi = 0.15$ was used to present the results of both models.

5.8 Maxwell's model

The classical model for the effective conductivity of dispersed particles is Maxwell's model, a theory that was originally developed to evaluate the conduction in heterogeneous media (Maxwell). The following equation, which is rewritten from equation 5.35, is the result of Maxwell's analysis:

$$\frac{k_{eff}}{k_f} = 1 + \frac{3\phi(r-1)}{2+r-\phi(r-1)}$$
(5.54)



Figure 5.13: A diagram of a solid particle surrounded by a fluid.

The approach of one dimensional heat conductivity was described above. Another approach is the mathematical treatment of Maxwell's theory when applied on thermal conductivity. Reviewing Maxwell's theory of effective thermal conductivity is important to show the differences in the assumptions and draw a comparison between both approaches. In an attempt to study the effect of particles in a medium, a starting point is to ask about the temperature field around a solid particle immersed in a fluid, as shown in Figure 5.13. The derivation of Maxwell's model is detailed in Appendix B.

Figure 5.14 shows the resulting equations from thermal resistance model 1, model 2, and Maxwell's model. A major difference between Maxwell's theory and the models developed in this work is that Maxwell's model is based on volume mixtures of particles and containing medium. The derivation of Maxwell's theory above showed that it assumes that the particles are far from each other and that the particle surrounding the temperature gradient is not affected by other particles.



Figure 5.14: A comparison of the effective thermal conductivity equations resulting from models 1, 2, and Maxwell's theory.

5.9 Conclusions

A single droplet evaporation model was developed and validated against experimental results. The development of this model was a step on a road map to understanding the effect of particles suspended on the surface of a droplet. Furthermore, the effective thermal conductivity was studied by conducting a thermal resistance analysis. The thermal resistance models were compared to Maxwell's model, which was derived from first principles based on heat transfer theory. The thermal resistance models showed that the particle thermal conductivity becomes influential when the evaporating droplet gets smaller. This work can be extended by studying the surface and the area ratios of particles and fluid contribution. Furthermore, a link has to be established between the thermal resistance model parameters and the results of the area contribution analysis. Finally, these models can be incorporated with the evaporation model.

Validating the developed model, which considers the effects of particles suspended on the surface of a droplet, can be achieved by employing experimental approaches that control and measure particle shape, thermal properties, and concentration within the droplet. Accurate temperature profiles of the evaporating droplet can be obtained using advanced techniques like thermal interferometry and infrared thermography. Reasonable assumptions may be made based on existing literature or experimental data when all parameters are not readily available. The model's accuracy and reliability can be assessed by comparing the experimental results with the model's predictions, considering the influence of the thermal resistance model parameters and the area contribution analysis results.

Chapter 6

Water Droplet Impact on Burning Wood Surfaces

In order to explore the mechanism of fuel surface cooling with water, an experiment was designed, and tests were conducted successfully for a water droplet impacting on surfaces of burning wood. This droplet impact condition was investigated experimentally for the first time. The main focus of this current investigation is to provide a behaviour description of a water droplet impacting a burning solid surface, with the intent to clarify the differences between impingement on cold surfaces and burning real application materials such as wood. Wood was used in the tests to simulate the temperature and surface characteristics of burning specimens. Sizes of water drops were in a range between 0.95 mm and 2.1 mm, resulting in a Weber number from 52 to 264. Visualisation of impact proved to be a challenge because of the optical characteristics of wood undergoing pyrolysis, the required high shutter speed and the very small droplet size. Images were improved significantly and used for quantitative measurements through careful post-processing and the application of a sequence of enhancement techniques. A new mechanism of fire extinguishment is proposed based on the sealant effect of the water liquid layer. The results showed a clear divergence in the behaviour of water splash on burning wood compared to cold surfaces. Prompt splash, receding breakup, and deposition were observed to occur at the initial stage of the droplet impact. The critical Weber number at which the impact drop breaks up and splashes with sub-droplets was shown to be lower for the tests on burning surfaces. Different types of wood also showed the dependence of droplet dynamics. Furthermore, the increase in droplet impact velocity revealed that most of the wetting was achieved during the first few milliseconds after impact and resulted in faster evaporation. Finally, a conceptual model is presented to summarise the behaviour of a water droplet impact on wood surfaces.

6.1 Introduction

Various fire extinguishment mechanisms have been described relevant to hydrocarbon fires including wetting and fuel surface cooling [37]. Investigations regarding surface cooling are targeted toward the effective use of water sprinklers and mist systems. Most importantly is the interaction between water droplets and fuel surface. Numerous studies on droplets surface impingement have been performed. The experimental investigations usually involve the impact of water droplets on a heated surface or a liquid film. Relevant studies on wood were conducted on room condition surfaces [38], [171], and heated surfaces [40]. Remarkably, the assessment of droplet impingement on a burning wood surface is lacking, which is investigated for the first time in this study to understand the first stage of fire extinguishment by water-based systems.

Whether a water-based suppression system can quench a fire depends on many factors that are usually studied such as droplet size distribution, spray location, nozzle operating pressure, the compartment geometry, and fuel type [72]. The present paper focuses on the local water surface interaction and dynamics, which play a major role in the heat exchange and mass transfer on the solid fuel bed.

Sprinkler fire suppression usually involves water droplet sizes in the order of 1 mm [172]. Droplets of this size are considered large in comparison to the optimal size of microns for water mist-based systems [9]. Large droplets penetrate fire plume without much contribution to the cooling of the gas phase. This is mainly due to droplet size and short residence time, which results in minor vaporisation. Only when the droplet reaches the fuel surface, fire suppression is then dominated by the interaction between liquid water and burning solid [173]. Different conditions apply when water droplets interact with a high-temperature gas phase environment or heated wall. Mainly, the gas phase environment involves mechanisms of heating, namely convective and radiant heating. However, conductive heat transfer is substantial for a droplet impacting on a hot surface. Generally, a droplet impinging on a solid surface might follow various scenarios or a combination of deposition on the fuel surface, absorption, splashing, forming a hot vapour layer at the interface, and rebounding [174], [31]. This results in a change in the behaviour of water flow on the surface which is influenced not only by the parameters of the drop but also by the properties of the target materials.

Water impact on organic materials characterised by the roughness due to their grooves has been rarely reported in the literature. Wood with higher density was correlated with lower critical impact Weber number for droplet splashing [171]. Furthermore, Lan et al. [40] tested water impact on a heated wood surface and observed liquid phase deformation at the solid wall due to the heat transfer between the solid and the liquid drop. Properties of splashed droplets over processed materials such as cardboard were also investigated [154]. Previously, no studies have focused on the dynamic aspect of splashes on burning natural woods and the outcomes of the process, which is important for the fire extinguishing process of wood constructions.

These outcomes are influenced by several factors involving both droplet and surface characteristics. The factors include liquid properties such as density, surface tension, droplet diameter, impact velocity and direction. In addition, surface features such as wettability, roughness, and temperature play a major role in the process [31]. The phenomena associated with all these properties require a restriction of variables when designing an experiment.

The current investigation aimed at providing a behaviour description of a water droplet impacting a surface of burning wood, with the intent to clarify the differences between impingement on cold surfaces and burning real application materials. Having thoroughly reviewed relevant journal papers to the current study, the following section provides a full description of the experimental procedure.

6.2 Methodology

6.2.1 Ignition procedure

The wood types used in this experiment were Lime, Beech, and Balsa, with a measured density of 606, 735, and 202 kg/ m^3 , respectively. The size of the samples was 15 × 15 mm and 5 mm in thickness. Each sample was ignited by heating the lower surface using a butane torch. Samples were heated until the flames could be sustained by the sample for a maximum of 25 seconds. Due to the relatively small size of the samples, the heat loss is significant to the surroundings compared to the released heat. Wood flames could only be sustained by the preheating the wood samples initially. Thus, the sample was put on a stand ready for the impact process explained below.

Figure 6.1 shows an image of a burning wood sample. The maximum flame height of nine samples is shown in the table. A 0.3 mm k-type thermocouple placed at the centre of the wood surface indicated that the maximum temperature varied with wood types.



Figure 6.1: A direct image of wood flame, flame heights and surface temperature ranges.

6.2.2 Droplet generation

The liquid droplets were generated using the droplet generator which was described in Chapter 3. Water is injected through a 0.5 mm nozzle for periods between 5 to 25 milliseconds. The droplet size (Do) range was between 0.95 and 2.1 mm with a step change of 0.2 mm approximately. The maximum variability was experienced in the smallest droplets with a percentage error of 10%. A droplet was formed as the water was ejected at the tip of the nozzle; the initial speed of droplets was a function of injection time. Although the initial speed of droplets varied from 0.2 to 1.6 m/s, the impact speed was controlled by changing the nozzle to the wood surface distance. Provided that droplets did not reach their terminal velocities, their speed is a function of the acceleration due to gravity and the fall distance.

6.2.3 Impact velocity

The nozzle height above the surface of the fuel was changed to produce impact velocities V_o of 2 and 3 m/s. The measured percentage error of impact velocities was below 4%. The velocities and droplet sizes were measured by imaging the droplet just before impact. The velocities were mapped with the settings of the generator such as ejection time and pressure level of the water. The Weber number varied between 50 to 293 assuming a constant surface tension, initial droplet diameter, and density.

The wettability of wood is significantly different from that of other materials usually studied in the literature such as metals and ceramics. Additionally, the roughness of the wood surface is expected to be different not only because of their origin and type of manufacturing process but also because of any post-process or coatings.

A small wood cube of a 15 mm side was used in this experiment. This size was minimised to reduce the variability of burning behaviour, but large enough to show the splashing behaviour.

6.2.4 Data acquisition and camera settings

A high-speed camera was used to observe the impingement evolution. The camera captured images at 10000 frames/s and exposure time of 1/10000s. Image resolution was 768×512 with a scale of 0.037 mm per pixel which resulted in a physical camera image size of 28.4×18.9 mm. During the experiment, the digital camera focused on the impingement location with the camera orientated at an angle to the horizontal axis.

The camera recording setting resulted in a total imaging time of 1.45 seconds. High frame rate setting was required to capture the dynamics of a droplet impact and have insightful details of liquid phase splash and evaporation. The recording mode was continuous, and an end trigger switch was used to stop the recording. The camera keeps recording the past 1.45 seconds continuously, only when the trigger button is pushed the recording stops. The end trigger was initiated manually 500-700 milliseconds after the droplet impact. Approximately half of the recording period was allowed for variation in the trigger time. Ultimately, all the recordings were synchronised to the impact moment frame. Each imaging series included the process of the droplets approaching the wood surface, impact, splash, secondary droplet, deposition, and initial evaporation. The camera was orientated at an angle to the horizontal on the sample surface. This orientation overcomes the difficulties of the limited depth of field in focus and the uneven burning wood surface. The part of the image in focus was set to the middle part of the sample surface where droplets were expected to land. Different from other techniques [35] [170], backlighting and direct imaging, in which the camera is orientated at the horizontal or at least situated just above the surface, of the impact cannot be utilised in this experiment. The reason is that burning wood surfaces are nearly always not balanced and the local variation of surface fluctuations can overshadow the droplet dynamics.

The experiment was not repeated at the same conditions of droplet size and impact speed. Rather, a total of 60 tests were conducted to examine the phenomenological aspects through step changes of droplet size. The examination of consecutive cases produced similar results. For example, the impact of droplets with diameters of $0.95~\mathrm{mm}$ and $1.12~\mathrm{mm}$ resulted in a maximum spread area increase of 3% with both splashes exhibiting no sub-droplets.

Significant spatial and temporal variation in temperature and surface characteristics meant that the acquisition of the temperature was extremely difficult even when different techniques are considered. The dynamic behaviour of the burning surface makes the placement of thermocouples impractical. Other methods such as thermal imaging face the issue of flame interference and the determination of burning surface emissivity. Moreover, surface features change dramatically because of the burning of the samples; a pre-test measurement of the roughness cannot be an indication of the surface condition in the time scale relevant to the impact.

6.3 Results and discussion

6.3.1 Visualisation and spread behaviour

Figure 6.2 shows the time evolution of a droplet impact on wood surfaces. The droplet diameter was 1.6 mm and has an impact velocity of 2 m/s. Figure 6.2 (a) and (b) are sequences of deformation behaviour upon a cold and burning Limewood surface, respectively. The cold case shows that the droplet spread to its maximum and receded to a smaller size without wetting the surface initially. On the other hand, the impact on the burning Lime surface resulted in secondary droplets with distribution from small to large sizes. The lamella maintained the maximum size and surface wetting was observed immediately. Interestingly, prompt splash followed by receding breakup occurred in addition to the liquid deposition, as shown in Figure 6.2 (b). Prompt splash occurs on rough surfaces and it involves droplet generations at the line of contact at the spread phase beginning when the lamella has a high radial velocity and is influenced by the structure of the surface. Receding breakup is a phenomenon of pure wetting, as the liquid retracts from its maximum spreading radius, droplets are left behind [221]. The general understanding is that these outcomes are to occur independently during the kinematic stage of the droplet spread [174].

As time passed, bubbles started to appear, a combination of fuel vapour escaping the surface and water boiling. The water droplet deformation and evaporation behaviour represent a general case for spreading on burning wood surfaces. However, droplet splash and the number of secondary droplets were significantly different and are discussed further below.



Figure 6.2: Images of the time evolution of droplets with 1.61mm initial diameter and impact velocity 2 m/s on Limewood and Beechwood: (a) cold Lime; (b) burning Lime; (c) cold Beech; (d) burning Beech.

Figure 6.2 (c) and (d) is the time elapsed images of droplet deformation on Beechwood. The droplet splash at 1.4 ms shows almost regular shape on the boundary of the liquid layer. Secondary droplets are considerably lower in this case. Evaporation was observed in the later stage of the impact.

The change in spreading behaviour suggests that it originates either from the surface or droplet characteristics. The surface of the burning wood becomes uneven and of elevated temperatures of at least 270°C and 400°C [43] [46]. Cracks are formed in the char which allows volatiles to escape easily [64]. Droplet surface tension can also change because of the deposition of soot and smoke particles on the surface [27] [204].

The immediate wetting and deposition on burning wood surfaces show that the liquid layer can behave as a sealant to the vapour of volatiles resulting from the wood pyrolysis. The water spreads along the cracks of wood which is known to be the easiest escape of volatiles [43]. This blocking effect can extend even when the heat is being transferred from the hot surface and flame radiation to the liquid. The evaporation stage of droplets shows that bubble can restrict the convection of fuel gases as well. The number of bubbles grows considerably as shown during the last stage in Figure 6.2 (d). Bubbles grow in size before bursting as a result of surface tension and they can hold a substantial amount of gases that would have ended up in the reaction zone otherwise. This mechanism is suggested to play a major role in the overall fire extinguishment in addition to the cooling of fuel surface. Furthermore, the continuous heat transfer from the surface to the water can lead to different boiling regimes. Descriptions of the several regimes are fairly complex because the various phenomena observed at impact must be addressed within each boiling regime [222]. This study main focus is on the phenomenological and morphological aspects of the impact and no details on the heat and mass transfer are discussed.

6.3.2 Initial stage and long-term evolution

Figure 6.3 presents a quantitative measurement of the observed spread factor of a 1.24 mm droplet impact on Limewood. For better comprehension of the general behaviour of the droplet, the short term evolution is plotted on Figure 6.3 (a), and the long-term progress on Figure 6.3 (b). The main divergence of the spread factor is realised after droplets had reached their maximum spread. The liquid on the burning surface continued to spread and, in most cases, flowed over the grooves of the surface creating an area of clear edges and angles. Then, the liquid layer started to evaporate and finally disappeared. On the other hand, the droplet on the cold surface receded to a minimum after the initial maximum spread. The droplet spreads gradually on the surface, noting that the liquid maintained certain contact angles and did not follow the texture streams on the surface.

The behaviour of droplet impacts on burning surfaces found in this study is different from the behaviour on heated surfaces described in the literature [4]. Both burning and heated surfaces share uncontrollable conditions when tested such as moisture content and undergoing pyrolysis. Furthermore, an experimental comparison between impacts on burning and preheating zones of a flame spread regime may lead to further insights.



Figure 6.3: The spread factor evolution for a droplet impact on Limewood with 1.24 mm initial diameter and 3 m/s impact velocity. The numbers on the images refer to the status indicated by the same numbers on the evolution profiles.



Figure 6.4: Spread evolution on of a droplet on burning Beechwood surfaces with different Weber numbers. (a) Droplet $D_o = 2.1mm$ impact on Beech burning surface. (b) Droplet $D_o = 1.2mm$ impact on Beech burning surface.

6.3.3 Effect of Weber number

Figure 6.4 is a comparison of two droplets impacting Beechwood burning surface at two speeds 2 m/s and 3 m/s. Figure 6.4 (a) shows the spread of 2.1 mm droplet which is higher than that of 1.2 mm droplet shown in Figure 6.4 (b). This is due to the increase in volume and the impact kinetic energy. When the impact speed was increased, both sizes wetted larger areas. As a consequence, the larger spread resulted in a faster rate of evaporation and absorption into the surface.

The increase of Weber number indicated a change in the boundary between the kinetic and deposition stages. The usual spread achieved in the deposition stage has moved to the first few milliseconds in the case of $W_e = 264$ as indicated in Figure 6.4 (a). While droplets with lower impact Weber numbers followed the general behaviour described above in Section 6.3.1.



Figure 6.5: Spread factor over time of droplets with 0.95mm initial diameter and 2m/s impact velocity on (a) burning surfaces, and (b) cold surfaces.

6.3.4 Wood density and texture

To describe the effect of wood density on droplet behaviour; Figure 6.5 shows the spreading of a 0.95 mm droplet with an impact velocity of 2 m/s. The lowest Weber number was chosen to minimise the fluctuations emerging from emitted secondary droplets. The three types of wood with different densities revealed different splashing behaviour. Surfaces of high-density wood resulted in the lowest liquid layer irregularities. In addition, lower density wood exhibited the shortest time to reach the maximum spread and more secondary droplets were observed. Generally, the average time for maximum spread and receding time appeared to increase on burning surfaces.

The critical Weber number was found to increase with density, in contradiction to the results in [171]. This might suggest that the critical number cannot only be compared to different density wood surfaces, but rather additional factors such as surface topography and droplet surface properties should be investigated. Although surface topography might affect the evaporation of droplets negatively since surface roughness increase resulted in less and larger secondary droplets [31], the secondary droplets in this investigation increased on wood surfaces of less density. Basic density of wood can also influence the results in addition to the surface roughness which resulted in discrepancies between models and experimental data [179].

6.3.5 Conceptual model

Figure 6.6 presents a conceptual model of a water droplet impact wood surfaces. Figure 6.6 (a) shows the impact on a cold wood surface where the droplet reaches a maximum spread factor initially and then exhibits a recoiling wave. The droplet then undergoes slow-spreading over the surface. Figure 6.6 (b) identifies the main conclusions on a droplet impacting a burning wood surface. The droplet achieves a higher spread factor compared to cold surface cases. The droplet wets the surface immediately as it spreads with secondary droplet formed during the same time as lamella expansion and following receding wave. The liquid then spreads along with the wood grains and groves. Bubbling is then observed and droplet evaporation continuous until complete disappearance.



Figure 6.6: Droplet impact behaviour conceptual model on (a) cold wood surface, and (b) burning wood surface.

6.4 Conclusions

An experimental study was designed and performed successfully for water droplet impact on burning wood surfaces. High-speed imaging with focused lighting was used to capture droplet burning surface images for the first time. Sufficient quantitative measurements showed the influence on the droplet spread and lifetime. The following conclusions can be drawn:

- 1. The observed maximum droplet spread on the burning surface tends to be larger than the cold surfaces which indicate a change in the characteristics of droplets and target surfaces.
- 2. Immediate wetting of burning surfaces and subsequent bubbling is suggested to play a role in preventing the escape of volatiles to the reaction zone. Hence, a local mechanism based on the sealant effect of water liquid layer is proposed in addition to the cooling of fuel surface.
- 3. The wood type significantly changes the behaviour of drop splash. Wood density increase was correlated with a more stable splash and a lower number of secondary droplets.
- 4. The critical Weber number is lower for burning surfaces, with more irregularities observed as Weber number increases. Similar Weber numbers for droplets different in size produce a different splash, maximum dimensionless spread.
- 5. Droplet on cold wood surfaces tend to wet the surface slowly and the spread starts to increase after the first unstable period of the impact.
- 6. Droplet on burning wood wets the surface immediately as it spreads to its maximum size. Then evaporation happens at the same time as penetration into the fuel. Bubbling was observed to occur followed by a decrease of observed liquid on the surface.

Chapter 7

The Effect of Thermal and Surface Characteristics of Burning Wood on Water Droplet Evaporation

The mechanism of fuel surface cooling was investigated experimentally by characterising the thermal and surface features of burning wood impacted by a water droplet for the first time. The main aim of this study is to explore the effect of wood surface features on droplet spread and evaporation. A distilled water droplet with a diameter of 2 mm was applied by an on-demand droplet generator and impacted the top surface of a burning wood sample at a speed of 2 m/s. The process was captured by a visual camera, two thermal cameras, and an infrared sensor to evaluate droplet impact, droplet evaporation time, droplet temperature evolution, surface wetting, and surface temperature. Additionally, the surface roughness of wood samples was assessed using a non-contact optical method to study texture variation at different burning stages. The results of this study indicated that evaporation time for droplets of the same conditions varied depending on the burning stage of impacting wood samples. Three stages of droplet lifetime were observed: spreading, combined absorption and evaporation, and evaporation of absorbed water. At maximum spreading, the droplet experiences pinning, causing the contact area to remain constant until the depletion of the visible water layer on the surface. Rapid heating of droplets was observed after spreading on the surface. The evaporation of droplets was evidently affected by the variation of wood surface structures.

7.1 Introduction

Various fire extinguishment mechanisms of solid fuel fires by water have been described, including wetting and fuel surface cooling [37] [223] [224]. Surface wetting is a primary mechanism where both the trajectories and evaporation rate of droplets define the capability of water spray to control or suppress a fire [172]. Many studies have focused on the effective use of water sprinkler, and mist systems have been some of the most reliable and effective methods for fire suppression applications [140] [193]. These studies are well suited for understanding global behaviour in terms of applied water volume and heat exchange. Further insights can be obtained by investigating the process of a droplet impact on burning wood and the involved dynamics and physical processes that influence evaporation. Remarkably, assessment of surface thermal and textures characteristics of burning wood samples effect on water droplet evaporation is lacking, which is investigated in this study to understand the early stages of water based fire extinguishment systems.

Detailed analysis of the phenomenological and morphological aspects of droplet impact on cold and burning wood samples using high speed imaging was presented by Albadi et. al. [39] and detailed in Chapter 6. The work showed major droplet spread and evolution processes and the effect of droplet diameter and impact speed. However, limited information was provided on surface temperature, and no surface characteristics were defined. Therefore, examining evaporation rate, surface temperature, and effect of surface tomography on droplet spread can provide further insights into the interaction between droplets and combustible surfaces.

Temperature measurement and visualisation of a surface and impinging droplet can be achieved by different methods. However, the condition of investigated case usually dictates the available options. Thermocouples, smoke analysers, and load cells are frequently utilised to measure pointwise temperature, heat release rate, and mass loss [223]. Furthermore, imaging techniques can provide spatial and temporal information. For example, visual imaging has been used to trace flame height, pyrolysis propagation, and fire spread [225]. Thermal imaging has been used to provide measurements for surface temperature, smouldering spread [226], and wood ignition. Surface temperature of biomass pellets was obtained using phosphor thermometry [227]. Wood density and gas phase temperature was measured using 3D X-ray tomography which enabled analysis of grain structure effect on pyrolysis [228].

Surface roughness measurements of wood surfaces are necessary to assess the effect on droplet spread and later evaporation. Traditional techniques that use a

stylus to pass over a surface can experience mechanical filtering because of the fine features [229]. Moreover, stylus based method not only face a challenge of nonhomogeneous fibrous materials such as wood, but can also damage charred surfaces. Hence, a non-contact method was applied in this investigation which is described in the methodology section.

Describing the multiphase dynamics of the studied process in this work continues to be challenging, in part because the whole interaction occurs in a short time along with varying surface features during combustion. This investigation attempts, for the first time, to characterise the surface temperature and texture of wood effect on the evaporation of a water droplet, surface characteristics at different stages of burning, and support previous work [39] with additional information on surface conditions. The results of this investigation showed three droplet lifetime phases: spreading, absorption and evaporation, and evaporation of absorbed water. The droplet contact diameter remained constant after spreading. Spreading and absorption is suggested to be influenced by surface roughness. Surface features was found to vary depending on the burning stage. Variation in surface roughness, spreading and absorption patterns was found to influence the evaporation time substantially.

7.2 Methodology

The experimental setup for droplet impact and evaporation used in this investigation is shown in Figure 7.1 (a). The main components were a burner, droplet generator, a visual camera, two thermal cameras, an infrared thermal sensor, and an illuminator. The droplet generator was described in detail previously [230]. The visual camera was used for direct imaging, while the thermal cameras operated in short and longwavelength for temperature measurements spanning from room temperature up to 700 °C.

The wood types used in this experiment were Lime, Beech, and Balsa. The cubic wood samples had a side length of 15 mm. The samples were put on two wires 40 mm above the burner nozzle ready for ignition. The droplet impact experiment was conducted eight times per wood type, and a total of 12 samples were prepared for surface roughness measurements per wood type. The uncertainty of the measurements was based on standard deviation.



Figure 7.1: (a) Experimental setup to study wood surface temperature during droplet impact and prepare samples for surface texture evaluation. The setup shows a visual camera along with two thermal cameras, infrared sensor for average temperature, light source, and a burner for wood sample ignition. (b) Schematic diagram of the non-contact optical system for surface texture measurements.

7.2.1 Surface topography

Alicona InfiniteFocusSL was used for roughness measurements. Alicona is a noncontact optical 3D measurement system based on focus variation for scanning structured surfaces. Alicona utilises the small depth of focus of an optical system presented in Figure 7.1 (b), along with a precise vertical drive to capture multiple images at varying vertical distances. During this scan, only some regions are in focus. The sharply focused regions in the images are then converted into one full resolution 3D map. Optics 5xAX was used which enabled 3.61 mm lateral measurement and an area of 13.03 mm^2 in a single field scan. This system has not been applied to the characterisation of wood surfaces in any known literature. The measurements were taken for the entire surface of wood samples using a multifield scan for areas up to 350 mm^2 using vertical and lateral resolutions of 460 nm and 10 µm, respectively. Wood ignition procedure is explained next along with the preparation procedure for surface roughness measurements.

7.2.2 Ignition

Wood samples were ignited using a premixed methane-air flame. Methane and airflow rates were set to 0.35 l/min and 0.9 l/min, respectively. The premixed flame then continued for a specific duration so that the wood samples sustain the flames and continue burning. Three heating durations of 10, 20 and 30 seconds were tested.

The duration of 20 seconds was the most suitable heating time as the flames were sustained in all three wood types and did not break any of the wood samples. The heating duration of 10 seconds did not ignite beech wood, while 30 seconds duration led to balsa wood breaking into separate elements. Ultimately, the samples were ready for droplet impact. The details of droplet generation and impact are discussed below.

Additional wood samples were ignited for surface measurements. These samples were quenched after 20, 40, and 60 seconds following the ignition of the pilot flame. Once a sample reached the allowed burning duration, it was put on an Aluminium plate and covered by a suitable glass cap to suppress the flame. The top surface of the samples was then examined for surface characteristics.

7.2.3 Droplet size and impact velocity

The diameter of water droplets used in the experiment was 2 mm. The variability of droplet size was within ± 0.1 mm. Moreover, droplet impact velocity V_o of 2 m/s was tested. The droplet size is dependent on the droplet generator operation. In contrast, the droplet impact velocity is dependent on the generator and the height of the generator nozzle above the wood surface. Both the size and impact velocity of water droplets at the onset of wood surface were measured using direct imaging.

7.2.4 Visual and thermal imaging

A visual camera was utilised to capture the top surface of wood samples. The camera with a Laowa 100mm f2.8 2X lens resulted in a physical camera image size of 48×37 mm, a spatial resolution of 47 µm/pixel, and 316 pixels spanning through a 15 mm profile over the top surface of wood samples. Two thermal cameras were used in this investigation to capture the temperature range of interest. A short-wavelength infrared InGaAs camera covered the temperature range 300°C to 700°C, and a long-wave FLIR Lepton camera captured a lower range between room temperature to 380°C. The C12741-03 InGaAs camera is Peltier cooled with sensitivity in the near-infrared region from 0.95 to 1.7 µm. The camera resolution is 840 × 512 and it captures a continuous sequence of images. The camera was fit with a 1.49-1.5 µm bandpass filter to restrict the spectral sensitivity. A Navitar 100mm/f2.8 lens was used, resulting in a spatial resolution of 75 µm/pixel.

The FLIR Lepton 3.5 thermal camera uses an uncooled VOx microbolometer sensitive in wavelength between 8 and 14 μ m. It provides a measurement range

suitable for water. In testing, the camera was found to saturate at 380°C. Calibration data was acquired using a blackbody furnace, which demonstrated the camera output was accurate to $\pm 2^{\circ}$ C across the usable range. Automatic flat field correction (FFC) is achieved using an integrated shutter every 3 minutes to compensate for ambient drift.

An infrared radiation thermometer (IRT) was used to measure the average temperature over the area in which the thermal cameras were sighted; its field-of-view (FOV) filled the entire target. The IRT featured a Hamamatsu P13243-011MA InAsSb photodiode, sensitive over wavelengths between 2.5 and 5.3 µm, a conventional transimpedance amplifier (TIA) and a CaF lens. The output voltage of the IRT was logged using a Pico Technologies ADC-20 Precision Data Logger at a sampling rate of 60 ms. With the use of a lookup table generated from a standard three-point calibration using a blackbody reference furnace and Planck's Law, the voltage was subsequently converted to temperature. A black body furnace (LANDCAL R1500T) with emissivity of 0.99 was utilised as a radiation source for calibration and to determine the Size-of-Source Effect (SSE) of the thermal cameras as explained below.

The data acquisition was synchronised using a start trigger signal controlling the visual and InGaAs cameras. A timestamp from this measurement was used to synchronise the Lepton and IRT measurements.

7.2.5 Data analysis

Along with the visualisation of the temperature maps, quantitative measurements were obtained using visual and thermal images by image enhancement, transformation, and registration. Visual images of charring wood surfaces presented a challenge because of the low contrast and minimal reflected light. Along with the correct settings of short exposure time and light flooding, an appropriate sequence of filters and noise reduction kernels enabled the evaluation of the wetted area.

Images of all cameras and IRT sensor data were analysed using MATLAB. Algorithms of area segmentation were applied to find the spread factor. Temperature values were found by applying Sakuma–Hattori interpolation equation to thermal camera data [225]. Furthermore, temperature measurements obtained from the thermal cameras were corrected for the size-of-source effect [231] by employing the size of the wetted area and average surface IRT data.

Surface data obtained by Alicona was processed using MeasureSuite Version 5.3 and Gwyddion Version 2.60. Profile roughness was found in two main directions along and across the grains. In addition, the average surface height was evaluated for the samples, which takes into account the topology over an area and is independent of direction.

7.2.6 Thermal cameras and sensors radiometric measurements

A thermal camera collects radiation from the target object and the surroundings. The radiation of the surroundings is reflected on the target surface. Furthermore, part of the object's radiation and the reflection gets absorbed by the atmosphere. The atmosphere itself produces additional radiation. Some of these contributions are taken into consideration depending on the calibration technique.

Figure 7.2 is an illustration of the thermal contribution of the wood sample and surroundings. The figure also shows the radiation received by the thermal camera. The surrounding emission that reaches the surface is W_o , while the wood emission is W_{wood} . Taking into account both contributions, Equation 7.1 describes the radiation received by the camera W_{tot} . The atmosphere transmission was assumed to be very close to unity because the distance between the camera and the target object was negligible.



Figure 7.2: A schematic representation of the thermal contribution of a wood sample and the radiation collected by an infrared camera. The diagram shows the properties of both the wood sample and the atmosphere.

$$W_{\text{tot.}} = \underbrace{W_{\text{wood emission}}}_{W_{\text{wood}}} + \underbrace{W_o \left(1 - \varepsilon_{\text{wood}}\right)}^{\text{wood reflection}}$$
(7.1)

Figure 7.3 is an illustration similar to Figure 7.2 with the addition of a water layer in between the wood sample and the atmosphere. The water layer has three material properties of emissivity, transmission, and absorption coefficients. The atmosphere contribution is also shown. Equation 7.2 states all the contribution components as received by the thermal camera, $W_{tot.}$.



Figure 7.3: A schematic representation of the thermal contribution of a wood sample, surroundings, water layer, and the atmosphere. The diagram also shows the radiation received by a thermal camera.

$$W_{\text{tot.}} = \underbrace{W_{\text{atm.}}}_{\text{wood emission}} + \underbrace{W_{\text{water }\varepsilon_{\text{water }}\tau_{\text{atm.}}}}_{\text{wood emission}} + \underbrace{W_{o}\left(1 - \varepsilon_{\text{water }} - \tau_{\text{water }}\right)\tau_{\text{atm.}}}_{\text{wood reflection}} + \underbrace{W_{o}\left(1 - \varepsilon_{\text{wood }} - \tau_{\text{water }}\right)\tau_{\text{atm.}}}_{\text{wood reflection}}$$
(7.2)

Equation 7.2 can be simplified by applying the following assumptions:

1. The atmosphere transmittance is approximately unity $\tau_{atm.} \approx 1$. This is justified by the close distance between the camera and the target surface.

- 2. Water transmittance is negligible $\tau_{water} \approx 0$ for relatively thick layers. Previous research showed that water layers of 10 micros are considered optically thick [232].
- 3. A value of 0.95 was used as the emissivity of the burning wood. This value was previously determined for the emissivity of wood and coke [233].

7.2.7 Correction of size-of-source effect

An infrared camera collects radiation from a well-bounded zone in front of the camera. The size of the conical zone is defined by the anglular aperture of the camera and lens system. This zone intersection with the target body defines the field of view (FoV). In an ideal situation, the field of view is bounded by a sharp perimeter or boundary, and given that the target entirely covers the field of view, the signal on the camera is independent of the size of the target body and its surroundings. The camera sensor contains many pixels and the concept of the field of view applies to each individual pixel.

Nevertheless, several effects can cause radiation from the FoV to fall out a pixel or the detection element and other radiation from outside of the FoV to reach the pixel. These effects are known as the size-of-source effect (SSE) [234], [235]. Ultimately, the SSE results in the alteration of the detection element signal based on the size of the target. Figure 7.4 shows the signal of the Flir Lepton v2.5 camera targeted towards a black body at a constant temperature and varying viewing aperture. The camera output increased with larger aperture sizes. This trend slowed down starting from 8 mm aperture size approximately. Following this point, the increase was not as significant as for the smaller sizes.

Generally, three techniques are used to find the SSE in thermal detectors [236]. Firstly, the direct technique, the detector measures a radiation source by varying aperture size. The influence of the source size on the measured signal can then be observed. The second technique involves a black spot to block the radiation on the source. Similar to the first technique, different sized apertures around the black spot are used to infer the SSE. Thirdly, the scanning technique where the detector is scanned across the source. The accumulation of the signals with distance from the source centre gives an indication of the SSE.

The direct method was used to correct the values of the measured signal, S. A thermal detector was directed towards a black body circular source while varying the aperture size. The SSE function, $\sigma_D(R)$, was determined by finding the ratio of the



Figure 7.4: Camera output when viewing a black body furnace though different sized apertures. The camera output is measured in Counts which is directly proportional to the radiation received at the sensor.

measured detector output, $S_{meas}(R)$, for a specific aperture radius, R, to the output, $S_{meas}(R_{ref})$, measured at a reference aperture radius, R_{ref} . Therefore, the SSE can be written as:

$$\sigma_D(R) = \frac{S_{\text{meas}}(R)}{S_{\text{meas}}(R_{\text{ref}})}$$
(7.3)

Figure 7.5 shows the evaluation of Equation 7.3 for the camera output presented on Figure 7.4. The SSE function was found by normalising around aperture size of 14mm. This particular size was chosen because the viewing field during the experiments was estimated to be 14 mm taking into account the target object.

Now, a thermal detector output can be detected using the SSE function using:

$$S_{\rm cal}\left(T_0\right) = S_{\rm meas} \, \frac{\sigma_{\rm D}\left(R_{\rm cal}\right)}{\sigma_{\rm D}\left(R\right)} \tag{7.4}$$

where the reference radius, $R_r ef$, is equal to the calibration aperture size, R_{cal} , and S_{cal} (T_0) is the signal corrected to the calibration aperture.



Figure 7.5: The normalised size-of-source effect (SSE) function with aperture diameters. The function is normalised with the respect to 14mm aperture diameter.

The reference signal $S_{meas}(R_ref)$ at R_ref is usually measured at the maximum aperture. Theoretically, this leads to the aperture where the SSE no longer changes. But depending on the equipment limitation, this point may never be reached practically. This does not pose an issue for correcting the detector output, as long as the SEE has been evaluated for the sizes up to the maximum size designed to be measured experimentally. In other words, the reference size, R_{ref} , can be a calibration radius, R_{cal} , which represents the maximum size intended for temperature measurements.

The case in the experiment was represented by a region with a radiance temperature T_0 , surrounded by one annular constant radiance area at temperature T_1 , and negligible radiation around these two regions. The full solution for correcting the SSE, in this case, was given in [231] which stated:

$$[h]S_{\text{cal}}(T_0) = \frac{\sigma_{\text{D}}(R_{\text{cal}})S_{\text{meas}} - [\sigma_{\text{D}}(R_1) - \sigma_{\text{D}}(R_{\text{cav}})]S_{\text{cal}}(T_1)}{\sigma_{\text{D}}(R_{\text{cav}})}$$
(7.5)

The radiance of surrounding approximation where $S_{\text{cal}}(T_i) \approx S_{\text{meas },i}$ can be used since only a small fraction of the signal is used to correct the signal detected at the centre of the target. In other words, the measured output can be used instead of the SSE corrected detector output. Hence, Equation 7.5 can now be written in directly measurable quantities as:

$$S_{\text{cal}}(T_0) = \frac{\sigma_D(R_{\text{cal}}) S_{\text{meas}} - [\sigma_D(R_1) - \sigma_D(R_{\text{cav}})] S_{\text{meas},1}}{\sigma_D(R_{\text{cav}})}$$
(7.6)

Equation 7.6 enables the determination of the temperature T_0 from the aperture region output S_{meas} , measurement of the surrounding region $s_{meas,1}$, and knowledge of the SSE function of the radiation detector.

Figure 7.6 shows visual and thermal images of a burning wood sample during a water droplet impact. The visual image indicates the water impact region by the low contrast region on the top surface of the wood sample. At the same time, the edges of the wood are bright due to the advanced stage of burning. The thermal image illustrates the low and high regions of the temperature. The middle region of the top wood surface is the droplet impact location. The low temperature region is surrounded by a high temperature which includes the top surface of the wood and the side faces as well. The distinctive features of this particular system suggest that it can be represented by two region geometry for the purpose of SSE correction.



(a) Visual image

(b) Thermal image

Figure 7.6: Visual and thermal images of a burning wood sample recorded during a water droplet impact. The burning fuel piece can be represented by a system of two regions, the droplet impact location, and the surrounding hot region.

The geometry is presented in Figure 7.7 with two annular regions. The central region has a radius R_{cav} , and a measured signal S_{meas} which represents the droplet impact area. The outer region represents the surroundings of the droplet impact area with a radius of R_1 , and a measured signal of $S_{meas,1}$.



Figure 7.7: A diagram representing a central cavity aperture surrounded by a constant radiance region. The radiance temperatures T_0 , and T_1 are assumed to be constants throughout each region.

7.3 Results and discussion

7.3.1 Droplet evaporation

Evaporation of a water droplet on lime and balsa is the focus of the discussion as previous results indicated that the most differences in phenomenological behaviour occurred between these two types [39]. Figure 7.8 shows the evaporation time of a 2 mm water droplet on lime and balsa woods. The droplets were ejected 20 and 40 seconds after the ignition starting time and process was recorded just prior to impact until the end life of the wood burning. The evaporation time was determined using two different methods, one was based on the visual recordings and the other based on the thermal images. These resulted in two different values for each case which are indicated by the stacked columns in Figure 7.8. Using the visual recordings, the evaporation time is measured from the impact moment to the disappearance of the water layer on the wood top surface. While the thermal imaging method depends on the temperature of the impact point starting from the sudden drop in temperature at the impact moment and for the duration droplet elevated temperature remains constant. The end of evaporation in the thermal measurements is marked by the start of surface temperature recovery.

Focusing on the thermal method values shown in Figure 7.8, the evaporation time of a droplet on lime wood was found to decrease from 4.9 to 3.3 seconds after impacting wood surfaces 20 and 40 seconds after ignition, respectively. In contrast, droplet evaporation increased from 5.1 to 7.9 seconds when impacting balsa surfaces in the same burning lifetime as lime.

In all the cases shown in Figure 7.8, thermal method times are longer than that obtained from the visual images which indicate how fast the visible water layer was consumed. Furthermore, the difference between the visual and thermal times suggests that part of the droplets was absorbed. For example, the percentage increase in the case of lime and balsa at 40 seconds was 153% and 98%, respectively. The next section explores the visualisation of droplet impact and temperature variation.



Figure 7.8: Evaporation time of a water droplet on lime and balsa wood surfaces after 20 and 40 seconds from ignition starting time. The error bars represent one standard deviation.

7.3.2 Visualisation of droplet impact and temperature evolution

Figure 7.9 shows the average surface temperature and temperature progression at droplet impact location for (a) lime and (b) balsa wood surfaces. The time axis values in the Figure are shifted so that time zero indicates the droplet impact and spread. Snapshots of the visual and thermal recordings are also shown at three distinctive times indicated by the dashed vertical lines.

The average surface temperature T_s , as shown in Figure 7.9 (a) and (b) was between 400 °C and 500 °C before impact. This temperature was within 50 °C range



Figure 7.9: Evolution of average surface temperature T_s and temperature at droplet impact point T_d on (a) lime and (b) balsa surfaces. Three vertical lines are shown on each plot representing from left to right: droplet spread after impact, disappearance of water layer in visual images, and end of evaporation. Time is shifted to the onset of impact.

of the temperature in the upcoming impact centre T_d , approximately. The surface temperature drops immediately when the droplet impacts the surface. It then reaches a minimum before it increases after droplet evaporation.

The value of T_d decreased rapidly after impact, marked by the first vertical line, because it indicated the water droplet temperature. Droplet temperature increased in approximately one second close to the boiling point, as shown by the curved profile after time zero for both wood types. Then, the temperature of the droplet remains constant.

Following the spreading and surface wetting, the droplet partly diffuses into the wood because of the capillary forces. This is not shown by the visual camera images where the water layer on the surface is the only obvious part of the droplet spread.
The droplet spread diameter in the visual images has already reached a maximum prior to this. However, examining thermal images obtained from both the Lepton and InGaAs cameras revealed the spread and absorption of water into the wood. The absorption is very clear by the distinctive and fast motion of the wetted area boundaries.

The water layer on the top of the surface continued to be consumed either by evaporation or absorption. At some point, noted by the second vertical line in Figure 7.9 (a) and (b), the water layer became no longer distinguishable from the surroundings in the visual images. However, Ts reached a minimum around this point which indicated an increase in the low-temperature regions as a result of the absorbed water spread. At the same time, T_d was still constant at the droplet temperature which suggests that water had not evaporated completely.

The apparent complete evaporation indicated by visual imaging was followed by the evaporation of the absorbed water. The temperature T_d remained almost constant below the boiling point in the duration between the second and the last vertical lines in Figure 7.9. The spread area in the thermal images of the droplet was decreasing gradually towards the point of the impact from the maximum spread area. This also explains the difference on the visual and thermal evaporation times presented in Figure 7.8.

When a droplet hits a surface, it can either undergo absorption or adsorption. Absorption occurs when the droplet penetrates into the surface and forms a uniform layer inside the bulk material, whereas adsorption happens when the droplet adheres to the surface without penetrating into the bulk material.

Adsorption is exothermic, releasing heat, while absorption is endothermic and requires energy input. When a droplet impacts a wooden surface, it undergoes partial adsorption while evaporating from the surface in contact with the air. Once the droplet disappears from the visual image, the desorption process begins, which causes the temperature at the drop's surface to decrease due to its endothermic nature. However, the reason behind this temperature drop requires further investigation in future research.

7.3.3 Surface roughness evolution

The surface roughness of wood was evaluated to assess its variation at different burning stages. Figure 7.10 shows the typical 3D surface texture of lime and balsa samples. The figure includes surfaces at the initial state and after the ignition in steps of 20 seconds. A part of the images is highlighted by a colour map which illustrates the

Time (s)	Lime $(7 \ \mu m)$	Balsa (7 μ m)	Beech $(7 \ \mu m)$
0	11.6 ± 1.6	15.4 ± 1.0	9.0 ± 0.1
20	11.6 ± 0.3	12.4 ± 0.3	7.0 ± 0.4
40	9.5 ± 0.3	10.8 ± 0.9	8.4 ± 1.4
60	8.7 ± 0.5	13.6 ± 1.9	11.7 ± 2.2

Table 7.1: Surface height S_a of wood and its variation with burning time. The cited uncertainty values are one standard deviation of nine samples per wood type.

surface peaks and valleys. The dark blue colour represents the valleys of $-40 \ \mu m$ or lower while the dark red indicates the peaks of 40 μm and above.

The maps show a clear trend that the structure became smoother in the case of balsa at 20 seconds compared to the initial state. However, the surface experienced more variations due to the development of cracks.

Table 7.1 shows the average surface height S_a of all the wood types at the initial state and later stages of burning. The trend discussed above can be seen in the surface texture of balsa and beech. Lime would have followed the same trend after 60 seconds as the surface is expected to develop larger cracks. This local behaviour on the surface may result from the interaction between chemical and thermomechanical stresses. These stresses may lead to different deformation regimes, including shrinkage and expansion [237].

Figure 7.11 illustrates average profile height values R_a of selected profiles on the top surface of the samples. Each column in the figure shows the average roughness along and across grain orientation. Each group of columns shows the evolution of roughness from the initial states to three burning lifetimes. The error bars are based on standard deviation.

Balsa had the most uneven surface across grain followed by lime and beech with R_a values of 12.8 7 µm, 11 7 µm, and 7 µm, respectively. The same trend was observed when measurements were taken along the grain orientation.

Decreasing surface roughness in lime can lead to enhanced contact between the droplet and the hot surface. This can explain the decrease in evaporation time of the later stage of burning lime shown in Figure 7.8. Furthermore, an increase in surface roughness can lead to liquid film disruption and make it more prone to splashing [238]. This was the case when comparing the results of balsa and lime burning wood samples [39] and the fact that balsa has higher surface features all the time compared to lime as presented in table 7.1. Research on hydrophilic non-porous structured surfaces



Figure 7.10: Surface texture of lime (left) and balsa (right) woods at different burning stages: (a) initial state, (b) 20, (c) 40, and (d) 60 seconds after ignition starting point. The colour maps indicate the peaks and the valleys on the surface.



Figure 7.11: Surface roughness R_a values of three types of wood: lime, balsa, and beech at different burning stages. Each column shows across and along grain average roughness of that particular type of wood and burning stage. The images on the top right illustrates lime wood surface with size of 12×12 mm. The error bars represent one standard deviation.

reported an increase in evaporation due to the intensification of internal convection [239].

Droplet evaporation at 40 second impact on balsa shown in Figure 7.8 increased considerably compared to the impact at 20 seconds. At the same time, the roughness values decreased moderately. This may be a result of an increase in absorption tendency and slow recovery of surface temperature. The rate of temperature recovery is affected by the evaporated water film which can lead to a decrease in the evaporation rate [223]. The water tend to be pulled from hot to cold regions as a result of thermocapillary stresses [224]. Furthermore, charred wood surfaces become more hydrophobic by changing liquid water contact angle but not necessarily reduce water absorption [240]. Water surface tension becomes lower at elevated temperatures and that more porous media can increase the capillary absorption of water [241].

7.3.4 Conceptual model

A summary of the observations on the life evolution of a droplet after impacting a wood surface is presented in Figure 7.12. The figure illustrates three phases and the evolution of droplet spread, droplet temperature, absorption area, and absorbed mass. A comparison between visual and thermal imaging suggests that a droplet undergoes three phases: spreading, combined absorption and evaporation, and evaporation of absorbed water. The time scale of these phases differs in magnitude. For example, spreading occurs in a few milliseconds, while absorption may take seconds. After the onset of droplet impact, the droplet spreads on the surface to a maximum contact diameter. The dynamics of spreading has previously been discussed [39]. One of the important realisations was that the spread on burning wood surfaces showed a wetting behaviour. During this phase, minimal absorption is expected because of the entrapped air and fuel volatiles. New results in this investigation presented the variation in surface roughness of burning wood. A decrease in surface roughness is suggested to increase the wetting and absorption in the second phase.



Figure 7.12: Schematic representation of the evolution of a droplet after impact on wood surfaces over different stages: Spreading, combined absorption and evaporation, and evaporation of absorbed water.

Following spreading, the absorption phase is characterised by the heating of the droplet to a fairly constant temperature and the disappearance of the water layer. By the end of this phase, a minimum average surface temperature is reached as a result of the maximum spread of absorption area. Finally, the last phase is the evaporation of the remaining absorbed water. The temperature of the surface is maintained at

the same droplet temperature. The end of this phase is marked by the absorption area shrinkage and fast recovery of the surface temperature.

7.4 Conclusion

Thermal and surface roughness measurements were obtained for burning wood samples and surface interaction with water droplet impact and evaporation. Visual and thermal imaging was employed to characterise the interaction dynamics. Additionally, surface roughness evolution was investigated by employing a non-contact technique. Following the measurements and analysis detailed above, the conclusions below can be drawn:

- 1. Three phases of droplet lifetime were identified: spreading, combined absorption and evaporation, and evaporation of absorbed water.
- 2. The apparent disappearance of the water layer on the surfaces may not necessarily indicate complete evaporation. Water spread was observed in the thermal images, and the droplet temperature profile suggested further evaporation of absorbed water.
- 3. The surface texture of wood varies with different burning stages. Generally, roughness values decreased to a minimum as the fibres on the structure were consumed in the case of lime and balsa. The variations in the roughness resulted in different droplet spread factors on wood, which in turn affected the contact area and heat exchange.
- 4. Wood surfaces can become more even, allowing a greater contact area at the interface. This also enables vapour pockets to escape during droplet spreading.
- 5. Different deformation regimes, including shrinkage and expansion, lead to an increase in wood surface variation at the later burning stages.

Chapter 8 Summary and Future Work

The work and experimental investigations in this thesis focused on the microscopic droplet behaviour during transient and steady conditions in fire environments. The type of cases selected in this work was based on the practical use of water spray for extinguishment applications. In chapter 1, an introduction was given on water use for tackling fires, followed by a list of the current research scope and objectives.

A detailed literature review of fire dynamics and mechanisms of fire extinguishment was given in Chapter 2. Primary and secondary mechanisms were discussed, along with water droplet evaporation processes. Surface phenomena such as the effect of nanoparticles on droplet surfaces were discussed. In addition, the combustion of solid materials and cellulose fuels in particular was reviewed.

In chapter 3, the developed methodology was detailed to investigate droplet evaporation, droplet surface tension, and the interaction of a droplet impacting a burning wood surface. Specifically, four different setups have been utilised to study particle effect on droplet evaporation, the effect of particles on droplet surface tension, phenomenological aspects of droplet impact on burning wood surfaces, and to characterise the thermal and surface characteristics of wood samples impacted by a single water droplet.

Chapter 4 explored water droplet surface contaminated by smoke particles, which is most likely in a real firefighting scenario. Droplets were evaporated by external heating to show the effect on the primary four mechanisms of fire extinguishment. Unlike previous experimental investigations on water interaction with fire, which were conducted in large-scale setups, this chapter demonstrated the significance of smoke contaminants on water droplet evolution and surface tension.

Transient water droplets evaporation was modelled in Chapter 5. Explicit and implicit schemes were formulated to take into account the receding radius of an evaporating droplet. The chapter also explores the average thermal conductivity resulting from the introduction of solid particles to the gas-liquid interface.

Chapter 6 explores the mechanism of fuel surface cooling with water. This chapter aimed to provide a behaviour description of a water droplet impacting a surface of burning wood, with the intent to clarify the differences between impingement on nonburning surfaces and burning real application materials. An experiment was designed, and tests were conducted successfully for a water droplet impacting surfaces of burning wood.

The mechanism of fuel surface cooling was investigated further in Chapter 7. An experiment was designed to characterise the thermal and surface features of burning wood impacted by a water droplet for the first time. The main aim of this study was to explore the effect of wood surface features on droplet spread and evaporation.

8.1 Evaporative and Interfacial Characteristics of a Droplet with Suspended Particles

- 1. Water droplets evaporation has been shown to be affected by smoke particle contamination. In particular, the surface of water droplets was deposited with smoke particles to simulate the effect of using water in an actual fire extinguishment situation.
- 2. Quantitative measurements on evaporating rate showed that contaminated droplets had a higher rate of evaporation. The results were consistent with available research on nanoparticle effect in fluids. However, previous experimental research investigated the properties changes by volume mixing, while this study focused on surface contamination, which is most likely in a water spray fire suppression process.
- 3. Surface tension was found to be lower for surface-contaminated droplets. The droplet surface motion changes in a way where clusters of particle networks move together around the surface. As the droplet evaporated, the particle concentration increased on the surface, leading to agglomerated particles. The surface tension change can considerably influence spray characteristics in fire extinguishment processes.
- 4. Two methods of surface tension measurement were developed and applied: fibre suspension and pendant droplet. In the fibre suspension method, the droplet

centroid relative to the fibre was used to formulate a force balance of the droplet, which enabled the measurement of surface tension. In the second method, the outer profile of a pendant droplet was used to calculate the surface tension by a numerical solution.

5. Analysis of surface tension indicated that the droplet surface tension changes dramatically during the deposition stage. For example, the surface tension of water droplets decreased by almost half.

8.2 Droplet Evaporation and Effective Thermal Conductivity Modelling

- 1. A theoretical methodology was presented to explore the effect of suspended particles on the surface of droplets. The methodology included the steps to model the evaporation process of a single droplet, effective thermal conductivity, and the available area for evaporation.
- 2. The transient heating of the droplet was shown to be non-linear evolution of the squared droplet diameter D^2 during the first stage of droplet evaporation lifetime. Throughout this stage, the droplet evaporation rate increased while it went through gradual thermal expansion. The droplet heating time ended when the droplet reached a linear regression of evaporation rate. At the same time, the droplet temperature reached a steady temperature.
- 3. The addition of particles on the surface of droplets was evaluated by modelling the effective thermal conductivity. The model assumed that the heat transfer is one-dimensional. This analysis meant that different heat exchange paths could be analysed as the transfer can occur in parallel, in series, or a combination of both.
- 4. Maxwell's theory of effective conduction was reproduced for thermal conductivity. The model was based on dispersed spherical particles in a larger sphere of another medium. This model was found to have two main shortcomings when evaluating systems with particles on the surface. The first limitation was that the model assumes particles are distanat from each other, and the other assumption is that particles are dispersed in the volume of the containing medium.

5. Modelling the average thermal conductivity based on a single dimension assumption revealed that conductivity has a major effect at the end of the evaporation process when the fluid layer and particles become comparable.

8.3 Water droplet impact on burning wood surfaces

An experimental study was successfully designed and performed for water droplet impact on burning wood surfaces. High-speed imaging with focused lighting was used to capture droplet burning surface images for the first time. Sufficient quantitative measurements showed the influence on the droplet spread and lifetime. The following conclusions can be drawn:

- 1. The observed maximum droplet spread on the burning surface tends to be larger than the cold surfaces, indicating a change in the characteristics of droplets and target surfaces.
- 2. Immediate wetting of burning surfaces and subsequent bubbling is suggested to play a role in preventing the escape of volatiles to the reaction zone. Hence, a local mechanism based on the sealant effect of water liquid layer is proposed in addition to the cooling of fuel surface. Bubbles can act as a wall in the same way as the unfissured zone. Heat conduction is affected by a network of cracks that initiates on the surface at a particular heating stage. A convective flow of hot gasses can penetrate through the cracks on the surface. Therefore, volatiles are restrained by structure in the unfissured zone.
- 3. The wood type significantly changes the behaviour of drop splash. Wood density increase was correlated with a more stable splash and a lower number of secondary droplets. This observation indicated that different wood types are a significant reason for the droplet spread behaviour. Furthermore, the number of secondary droplets was different
- 4. The critical Weber number is lower for burning surfaces, with more irregularities observed as Weber number increases. Similar Weber numbers for droplets different in size produce a different splash and maximum dimensionless spread.
- 5. Droplets on cold wood surfaces tend to wet the surface slowly, and the spread starts to increase after the first unstable period of the impact.

6. Droplet on burning wood wets the surface immediately as it spreads to its maximum size. Then evaporation happens at the same time as penetration into the fuel. Bubbling was observed, followed by a decrease of observed liquid on the surface.

8.4 The effect of thermal and surface characteristics of burning wood on water droplet evaporation

Thermal and surface roughness measurements were obtained for burning wood samples and surface interaction with water droplet impact and evaporation. Visual and thermal imaging was employed to characterise the interaction dynamics. Additionally, surface roughness evolution was investigated by employing a non-contact technique. Following the measurements and analysis detailed above, the conclusions below can be drawn:

- 1. Three phases of droplet lifetime were identified: spreading, combined absorption and evaporation, and evaporation of absorbed water. The spreading is the first stage after the onset of impact until the droplet reaches a maximum spread. This was followed by combined absorption and evaporation. This phase was characterised by the heating of the droplet to a fairly constant temperature and the disappearance of the water layer on the surface, as was seen in the visual images.
- 2. The time scale of the three identified phases differs in magnitude. Spreading occurred in a few milliseconds, the combined evaporation and absorption phase took a second, approximately, and the last phase of evaporation lasted for a few seconds. The difference in magnitude shows that certain phases can be beneficial for enhancing extinguishment mechanisms. The initial spreading and combined phases may lead to a direct vapour layer between the fuel and flame. Information about the mass of water can show the different contributions of these three phases.
- 3. The apparent disappearance of the water layer on the surfaces may not necessarily indicate complete evaporation. Water spread was observed in the thermal images, and the droplet temperature profile suggested further evaporation

of absorbed water. The temperature profile at the location of droplet impact indicated temperatures below the boiling points of water.

- 4. The surface texture of wood varies with different burning stages. Generally, roughness values decreased to a minimum as the fibres on the structure were consumed in the case of lime and balsa. The variations in the roughness resulted in different droplet spread factors on wood, which in turn affected the contact area and heat exchange.
- 5. Wood surfaces can become more even, allowing a greater contact area at the interface. This also enables vapour pockets to escape during droplet spreading. A decrease in surface roughness can lead to enhanced contact between a droplet and hot surfaces. The contact area is directly linked to heat transfer and shorter evaporation times.
- 6. Different deformation regimes, including shrinkage and expansion, lead to an increase in wood surface variation at the later burning stages. In addition, the deformation affected the spreading of water droplets in the initial phases, and it can improve evaporation due to the intensification of internal convection.

8.5 Future work

The results and analysis discussed in this thesis lead to more research questions on the evaporation dynamics of water droplets. The following research subjects could expand this current investigation and may shed further light on the gap presented in this work.

8.5.1 Multi droplet system and interaction with smoke layer

Studying the interaction of a system of several droplets with smoke layer can provide better analysis of spray dynamics. Evaporating droplets affect the smoke field by diluting oxygen as a result of vapour production. The effect of convection during evaporation is expected to enhanced by the presence of particles on droplet surfaces. Furthermore, employing suitable imaging techniques can enable the visualisation and evaluation of particles agglomeration, droplet shape distortion, and effect on evaporation rate.

8.5.2 Water droplet modelling including the effect of particles on the gas liquid interface characteristics

A model to study the effect of smoke particles on the surface of evaporating water droplets could be established. Including the transient evaporation condition and the modelling of smoke particle geometry on the surface would enable a better explanation of the heat and mass transfers. Additional research in this area would provide the opportunity to predict and perhaps discover new techniques to increase the efficiency of water evaporation in fire extinguishment applications, particle interference, and enhanced atomisation features. In addition, the model can be validated by exploring methods to evaluate the concentration of suspended particles. The concentration of smoke can be controlled by designing a setup where a droplet is allowed to move through a container which contains smoke. Droplets can then be collected and dried. The particle concentration may be linked the residence time of droplets in the container.

8.5.3 Acoustic phenomena and the effect on droplet evaporation

This investigation would provide insights into the the interaction of the acoustic field on the evaporation process of water droplets. The research would include the acoustic waves influencing the distortion of droplet surface. In theory, the exposed droplets would have a larger surface area of the liquid to the ambient environment compared to spherical droplets. Increased surface area would benefit further exploration of water droplet and the air currents surrounding the distorted droplet surface. In addition, the coupled phenomena of heat and mass transfer in an acoustically disturbed droplet could be further explored in this investigation.

8.5.4 Investigation of the spread of flames under the suppression effect of water droplets using a synchronised multi-imaging system

Imaging the suppression behaviour of water droplets could show the effect of water droplets on the spread of cellulosic fuels. A synchronised multi-imaging system could include a high speed colour camera to capture the burning process, an shortwavelength infrared InGaAs camera for fuel surface temperature measurement, and a long-wavelength thermal camera for low temperature measurements around room temperature, and a high speed camera for Schlieren to visualise the flow field around the burning fuel. This study can directly provide quantitiave measurements on the burning lifetime, charring distance and speed, thermal visualisation and gas phase flow patterns. Furthermore, this study will show the different process related to both the solid and gas phases simultaneously in addition to the temperature measurements. The visualisation would show how the water evaporation affect the spread of the flames as the vapour leaves the surface to the enter the flames region. At the same time, the temperature fields could be evaluated to show the heat transfer as water droplets spread and evaporate over the surface.

8.5.5 Three dimensional investigation of fuel beds interaction with evaporating water

Visualisation of the water droplet impact in three dimensional images would enable further information on the evaporating liquid phase and the effect of different surfaces. The methodology would provide significant quantitative measurement on the number of nucleated bubbles of vapour in the droplet, their size, their speed of development, sub droplet ejection, elongated ligaments or liquid phase fingers during impact. The research can be extended to the assess water additives on the thermal behaviour of fuel. In certain cases, salty water may be used for fire extinguishment. These cases and their behaviour can be explored and compared with each other to explore their viability and efficiency. Furthermore, the surface of a burning fuel can be investigated to further understand the dynamics of disruptive process during droplet evaporation. Techniques such as stereo imaging, optical coherence tomography, and X-ray computed tomography could be utilised in this study. Appendices

Appendix A D-squared Law: Evaporation of a Single Droplet

Heat and mass transfer rates increase with larger surface area, a typical technique is to use water mist in flame extinguishment applications instead of continuous flow. Water droplets have more interfacial area than that of the original continuous flow of the same mass. The dispersion of a continuous flow into droplets is called atomisation or spraying. Therefore, understating droplet heat, mass, momentum exchange is necessary to understand spray behaviour.

D-squared law is derived by evaluating the mass loss rate due to evaporation. A droplet of radius r vaporises in an atmosphere of temperature T_{∞} and mass fraction w_{∞} . The droplet contains one species only and therefore the parameters refer to that species. The droplet is considered here to be water and the atmosphere is air.



Figure A.1: Transport phenomena on a droplet surface

Droplet evaporation models for a single component assume that the vapour is saturated at the surface of the droplet. This idea allows to close the problem which will be shown later in details. Hence, the evaporation process becomes a mass transfer problem from the droplet surface of saturated vapour to the ambient gas. The evaporation of a droplet placed in stagnant air involves two processes:

- Molecules escape from the surface to become in the gas phase just above the surface.
- The diffusion of these molecules into the ambient air.

It can be argued that in the molecular level no definite liquid surface can be defined. In fact, a transition layer with change in density is located between on the droplet.

The first process can be investigated in terms of molecular dynamics and can only be approached with certain assumptions for macroscopic theoretical studies. In combustion, researchers mainly paid attention to the second step of the evaporation.

A.1 Mass transfer

Generation of depletion of species at an interface induces a flowing fluid. This phenomenon is called Stefan's flow named after Stefan who first investigated an evaporating fluid in a tube. Examples of processes that produces or removes species include chemical reaction, condensation, evaporation, and absorption. Stefan original papers were in German and remained in the shadows for a very long time. An interesting fact is that Stefan's work was independently reproduced around a century later [242].

Distinction should be made between Stefan's flow and diffusion which is described by Fick's law. Addition or removal of species on a surface creates a net flow as the fluid next to the surface is displaced by the change in species amount on the surface. If concentration gradients are present, diffusion also takes place relative the flow. The total rate of mass transfer then becomes a contribution of both diffusion and Stefan's flow.

Taking an evaporating droplet in air as an example, vapour at the droplet surface moves away from the interface as it is displaced by additional vapour from the droplet. In this case, vapour is transported by convection while concentration gradient will also impose diffusive transport.

In general, there are two approaches for evaporation rate derivation based on mass transfer. The first approached followed in [243] which starts by the differential equations for species conservation. This approach requires imposing Stefan flow as a boundary condition. The second approached presented in [244] where the derivation starts by considering the mass flux surrounding the droplet. The second method is clearer in terms of visualising Stefan's flow. However, a discussion on the relevant assumption in not given in both references and others. An attempt is made here to derive the evaporation rate based on mass transfer using the second approach. All related assumptions and their implications are also discussed below.

Before dealing mathematically with the problem, it is important to review basic definitions. The partial density of species i is

$$\rho_i = m_i / V(\mathrm{kg}\,\mathrm{m}^{-3}) \tag{A.1}$$

The overall density can then be expressed as

$$\rho = m/V = \sum m_i/V = \sum \rho_i \tag{A.2}$$

Mass concentration can be written as the ratio of partial density to the total density

$$w_i = \frac{m_i}{m} = \frac{m_i/V}{m/V} = \frac{\rho_i}{\rho} \tag{A.3}$$

Where w_i is the mass fraction of species i

For a moving flow with concentration gradients, velocities of species can be viewed as a summation of flow bulk velocity and diffusion velocity. Figure 2 shows an evaporating droplet, denoting liquid vapor as species A, and air as species B, their respective velocities can be expressed as

$$V_A = V + V_{diff,A}$$

$$V_B = V + V_{diff,B}$$
(A.4)



Figure A.2: Diffusion of droplet with the assumption of stationary atmosphere



Figure A.3: Mass diffusion in addition to bulk transfer [245]

The microscopic representation just above the surface is presented on Figure 3. The rate of mass flow \dot{m} through an area A can then be stated as

$$\dot{m}_{A} = \rho_{A}V_{A}A = \rho_{A}(V + V_{diff,A})A = \rho_{A}VA + \rho_{A}V_{diff,A}A$$

$$= \dot{m}_{conv,A} + \dot{m}_{diff,A}$$

$$\dot{m}_{B} = \rho_{B}V_{B}A = \rho_{B}(V + V_{diff,B})A = \rho_{B}VA + \rho_{B}V_{diff,B}A$$

$$= \dot{m}_{conv,B} + \dot{m}_{diff,B}$$
(A.5)

The previous equations can also be written in terms of mass flux $j = \dot{m}/A$ as

$$j_{A} = \rho_{A}V + \rho_{A}V_{\text{diff},A} = \frac{\rho_{A}}{\rho}\rho V - \rho D_{AB}\frac{dw_{A}}{dx}$$

$$= w_{A}(j_{A} + j_{B}) - \rho D_{AB}\frac{dw_{A}}{dx}$$

$$j_{B} = \rho_{B}V + \rho_{B}V_{\text{diff},B} = \frac{\rho_{B}}{\rho}\rho V - \rho D_{BA}\frac{dw_{B}}{dx}$$

$$= w_{B}(j_{A} + j_{B}) - \rho D_{BA}\frac{dw_{B}}{dx}$$

(A.6)

At any point the total mass flow rate equals the summation of different species mass flow rates

$$\dot{m} = \dot{m}_A + \dot{m}_B \tag{A.7}$$

The total mass flow is $\dot{m} = \rho V A$

$$\rho VA = \rho_A V_A A + \rho_B V_B A \tag{A.8}$$

The average velocity of mass flow V can be expressed as

$$V = \frac{\rho_A V_A + \rho_B V_B}{\rho} = \frac{\rho_A}{\rho} V_A + \frac{\rho_B}{\rho} V_B = w_A V_A + w_B V_B \tag{A.9}$$

In fact, V is the velocity that would be measured if a sensor is placed in the flow.

To solve equation for water droplet evaporation, several assumptions are required. The mass flux equation can be written in terms of mole fraction concentration and the total molar density of gas vapor mixture can be assumed to be constant. This assumption requires the pressure and temperature of the mixture to be constant as explained in [245]. Another assumption is that total density is constant which is a commonly used approach [246]. But this implies that the molecular weights of mixture components are the same or the composition molecular weight is constant in the domain.

With the assumption of constant total density $\rho = \rho_A + \rho_B = \text{ constant}$, the equation for mass flux can be written as

$$j_A = w_A \left(j_A + j_B \right) - \rho D_{AB} \frac{dw_A}{dr} \tag{A.10}$$

It is assumed that air does not diffuse into water $j_B = 0$, this assumption implies that air is stationary and the diffusion velocity towards the droplet is opposed by the convective flow (Stefan's flow) away from the droplet.

$$j_A = w_A j_A - \rho D_{AB} \frac{dw_A}{dr} \tag{A.11}$$

For steady conditions, mass flow rate of vapour is assumed to be constant across r, note that the mass flux cannot be considered constant as the area changes with r

$$j_A = \frac{\dot{m}_A}{A} = \frac{\dot{m}_A}{4\pi r^2}$$

$$\dot{m}_A = j_A A = \rho_A V_A (4\pi r^2) = constant$$
(A.12)

Rearranging the equation, note that total density was assumed to be constant previously.

$$\frac{\dot{m}_A}{4\pi r^2} = -\frac{\rho D_{AB}}{1 - w_A} \frac{dw_A}{dr} \longrightarrow -\frac{4\pi r^2}{1 - w_A} \frac{dw_A}{dr} = \frac{\dot{m}_A}{\rho D_{AB}} = \text{ constant}$$
(A.13)

Since \dot{m}_A, D_{AB} , and ρ are constants, this is proceeded by separation of variables and integration of the equation with the boundary condition of

$$r = r_S, w_A(r = r_S) = w_{A,S}$$
 (A.14)

where r_s is radius of the droplet surface, and $w_{A,S}$ is the mass density fraction at the surface.

$$-\int_{w_{A,S}}^{w_{A}} \frac{dw_{A}}{1-w_{A}} = \int_{r_{S}}^{r} \frac{\dot{m}_{A}}{4\pi r^{2}\rho D_{AB}} dr$$
(A.15)

$$\ln\left(\frac{1-w_A}{1-w_{A,S}}\right) = \frac{\dot{m}_A}{4\pi\rho D_{AB}} \left(\frac{1}{r_S} - \frac{1}{r}\right) \tag{A.16}$$

Rearranging to find w_A

$$w_A(r) = 1 - (1 - w_{A,S}) e^{\left(\frac{\dot{m}_A}{4\pi\rho D_{AB}} \left(\frac{1}{r_S} - \frac{1}{r}\right)\right)}$$
(A.17)

Also, using the following condition of the mass density fraction far away from the droplet

$$r = r_{\infty}, w_A(r = r_{\infty}) = w_{A,\infty} \tag{A.18}$$

The evaporation rate can be expressed as

$$\dot{m}_A = 4\pi r_S \rho D_{AB} \ln\left(\frac{1 - w_{A,\infty}}{1 - w_{A,S}}\right) \tag{A.19}$$

The equation is commonly presented as

$$\dot{m}_A = 4\pi r_S \rho D_{AB} \ln\left(1 + B_M\right) \tag{A.20}$$

$$B_M = \frac{w_{A,S} - w_{A,\infty}}{1 - w_{A,S}}$$
(A.21)

where B_M is called Spalding mass transfer number.

A.2 Heat transfer

The derivation of evaporation rate above was based on mass transfer of vapour above the droplet surface. The problem can be also described in terms of heat transfer. When steady conditions are reached, heat supplied by the surrounding to the droplet results in evaporation. Similar to mass transfer based derivation, the problem can be solved either by considering the heat transfer differential equation [243], [244], [247] , or by writing the heat flux balance surrounding the droplet [246]. Both methods are equivalent, the second approach in fact is a result of the first after one step of integration and imposing the relevant boundary conditions. Here the differential equation approach is presented.

$$\frac{\dot{m}c_{pg}}{4\pi k}\frac{dT}{dr} = \frac{d}{dr}\left(r^2\frac{dT}{dr}\right) \tag{A.22}$$

Where c_{pg} is the specific heat of the gas phase, and k is the thermal conductivity of the vapour mixture. The boundary conditions are the surface temperature and the ambient temperature far away from the droplet are

$$T(r = r_s) = T_s$$

$$T(r \longrightarrow r_{\infty}) = T_{\infty}$$
(A.23)

$$T(r) = \frac{(T_{\infty} - T_s) e^{(-Z\dot{m}/r)} - T_{\infty} e^{(-Z\dot{m}/r_s)} + T_s}{1 - e^{(-Z\dot{m}/r_s)}}$$

$$Z = \frac{c_{pg}}{4\pi k}$$
(A.24)

At the boundary, the heat supplied by conduction equals the heat used in the evaporation process

$$\dot{m}h_{fg} = 4\pi k r_s^2 \left[\frac{dT}{dr}\right]_{r_s} \tag{A.25}$$

Differentiating temperature domain equation and using the above boundary condition, the evaporation rate can be expressed as

$$\dot{m} = \frac{4\pi k r_s}{c_{pg}} \ln\left(1 + \frac{c_{pg}\left(T_{\infty} - T_s\right)}{h_{fg}}\right) \tag{A.26}$$

The equation is commonly presented as

$$\dot{m} = \frac{4\pi k r_s}{c_{pg}} \ln \left(1 + B_T\right)$$
 (A.27)

$$B_T = \frac{c_{pg} \left(T_{\infty} - T_s\right)}{h_{fg}} \tag{A.28}$$

Appendix B Maxwell's model

The classical model for the effective conductivity of dispersed particles is Maxwell's model, a theory that was originally developed to evaluate the conduction in heterogeneous media (Maxwell), the following equation is the result of Maxwell's analysis:

$$k_{eff} = k_f \frac{2k_f + k_p + 2\phi \left(k_p - k_f\right)}{2k_f + k_p - \phi \left(k_p - k_f\right)}$$
(B.1)

This can be rewritten in the form

$$\frac{k_{eff}}{k_f} = 1 + \frac{3\phi \left(r - 1\right)}{2 + r - \phi \left(r - 1\right)} \tag{B.2}$$



Figure B.1: A diagram of a solid particle surrounded by a fluid.

The approach of one dimensional heat conductivity was described above. Another approach is the mathematical treatment of Maxwell's theory when applied on thermal conductivity. Reviewing Maxwell's theory of effective thermal conductivity is important to show the differences in the assumptions and draw a comparison between both approaches. In an attempt to study the effect of particles in a medium, a starting point is to ask about the temperature field around a solid particle immersed in a fluid, as shown in Figure B.1. Three variables are illustrated in Figure B.1, the radial axis r origin at the centre of the solid particle, the vertical axis z, and an angle θ which describes the rotation of the radial axis. The radius of the particles is given by r = a. The temperature differential equation is given by:

$$\frac{1}{r^2}\frac{\partial}{\partial r}\left(r^2\frac{\partial T}{\partial r}\right) + \frac{1}{r^2\sin\theta}\frac{\partial}{\partial\theta}\left(\sin\theta\frac{dT}{\partial\theta}\right) = 0 \tag{B.3}$$

This is a second order differential equation which can be solved by the separation of variables. The assumption is that the temperature is a result of two functions, one that depends on r, and the other depends on θ only. This assumption can be stated as:

$$T(r,\theta) = R(r)\Theta(\theta)$$
(B.4)

Substituting Equation B.4 into Equation B.3 results in the following:

$$\Theta\left(\theta\right)\frac{1}{r^{2}}\frac{d}{dr}\left(r^{2}\frac{dR\left(r\right)}{dr}\right) + \frac{R\left(r\right)}{r^{2}\sin\theta}\frac{d}{d\theta}\left(\sin\theta\frac{\Theta\left(\theta\right)}{d\theta}\right) = 0 \tag{B.5}$$

Multiplying by $\frac{r^2}{R(r)\Theta(\theta)}$:

$$\frac{1}{R(r)}\frac{d}{dr}\left(r^{2}\frac{dR(r)}{dr}\right) + \frac{1}{\Theta(\theta)\sin\theta}\frac{d}{d\theta}\left(\sin\theta\frac{\Theta(\theta)}{d\theta}\right) = 0$$
(B.6)

Equation B.6 presents a remarkable outcome of the separation of variables step. The first term depends on r only, and the second term depends on θ only. Hence, both terms must equal to a constant for this equation to be true. Let both terms equal to k and -k. respectively:

$$\frac{1}{R(r)}\frac{d}{dr}\left(r^{2}\frac{dR(r)}{dr}\right) = k \tag{B.7}$$

$$\frac{1}{\Theta(\theta)\sin\theta}\frac{d}{d\theta}\left(\sin\theta\frac{\Theta(\theta)}{d\theta}\right) = -k \tag{B.8}$$

These equations are ordinary differential equations (ODEs) which can be solved individually. Expand the ODEs using the chain rule

$$r^{2}\frac{d^{2}R(r)}{dr^{2}} + 2r\frac{dR(r)}{dr} - kR(r) = 0$$

$$\frac{d^{2}\Theta(\theta)}{d\theta^{2}} + \frac{\cos\theta}{\sin\theta}\frac{d\Theta(\theta)}{dr} + k\Theta(\theta) = 0$$
(B.9)

Setting k = l(l+1), the ODE of $\Theta(\theta)$ can be transformed to

$$\frac{d^2\Theta\left(\theta\right)}{d\theta^2} + \frac{\cos\theta}{\sin\theta}\frac{d\Theta\left(\theta\right)}{dr} + l(l+1)\Theta\left(\theta\right) = 0 \tag{B.10}$$

This equation has a solution in terms of Legendre polynomials. Legendre polynomials are one kind of special functions which are met in the solution of many physical systems. Legendre polynomials are usually donated by $P_n(x)$, where n is the order of the polynomial.

$$\Theta\left(\theta\right) = C_l P_l\left(\cos\theta\right) \tag{B.11}$$

for l = 0, 1, 2, 3, ..., and where C_l is a constant that can depend on l, and $P_l(\cos \theta)$ is the Legendre polynomials.

On the other hand, the ODE of R(r) can be rewritten using the transformation of k = l(l+1) as:

$$r^{2}\frac{d^{2}R(r)}{dr^{2}} + 2r\frac{dR(r)}{dr} - l(l+1)R(r) = 0$$
(B.12)

This equation has the following general solution:

$$R(r) = A_l r^l + \frac{B_l}{r^{l+1}}$$
(B.13)

where A_l and B_l are constants. Using the solutions of the individual ODEs, the general solution of the temperature profile can then be written as:

$$T(r,\theta) = R(r)\Theta(\theta)$$

$$T(r,\theta) = \left(A_l r^l + \frac{B_l}{r^{l+1}}\right) \left(P_l(\cos\theta)\right)$$
(B.14)

The constants A_l and B_l may depend on l in the same way as the C_l which was encountered in Equation B.11. The constant C_l was impeded in A_l and B_l since the multiplication of constants results in other constants. Furthermore, l is an infinite number starting at zero, hence, the solution will be a sum over all values of l:

$$T(r,\theta) = \sum_{l=0}^{\infty} \left(A_l r^l + \frac{B_l}{r^{l+1}} \right) \left(P_l(\cos\theta) \right)$$
(B.15)

B.0.1 Boundary conditions

Figure B.2 is an illustration of the boundary conditions of the particle and fluid system. The first boundary condition is the symmetry at the centre of the particle, which is typical of spherical solids. The second boundary condition is related to the temperature field around the particle. The third and fourth conditions represent the temperature continuity and heat flux at the particle fluid interfaces, respectively.



Figure B.2: The boundary conditions of the system.

Mathematically, the first boundary condition (BC. 1) at r = 0 is

$$T_p|_{r \to 0} = const. \tag{B.16}$$

The second boundary condition (BC. 2) involves the temperature gradient as r approaches infinity. It can be stated in different expressions depending on the gradient form. For a linear change in temperature, the second boundary condition is:

$$T_f|_{r \to \infty} = bz$$

$$T_f|_{r \to \infty} = T_{\infty}z$$
(B.17)

The third boundary condition (BC. 3) is the temperature continuity at the particle fluid surface. This boundary condition implies that the contact resistance is negligible. BC.3 can be stated as follows:

$$T_p|_{r=a} = T_f|_{r=a} \tag{B.18}$$

The fourth boundary condition (BC. 4) is the continuity of the thermal flux between the different mediums. Note that each medium has a specific thermal conductivity. Therefore, the temperature gradients are different at each side of the interface, given the inequality of the thermal conductivity values. The last boundary condition expression is:

$$-k_p \frac{dT_p}{dr}\Big|_{r=a} = -k_f \frac{dT_{lr}}{dr}\Big|_{r=a}$$
(B.19)

B.0.2 Solution

The general solution of the temperature profile in Equation B.14 can be applied within the particle and the surrounding medium. And applying the boundary conditions requires making a distinction between the particle temperature field $0 \le r \le a$ and the field within the fluid medium $r \ge a$. By applying BC. 1 in Equation B.14, the second term with the radius in the denominator does not reflect the physics of the problem by implying that the temperature increases infinitely. As a consequence, the constant B_l must equal zero at all the values of l for the temperature to be finite. The temperature profile can then be written as:

$$T(r,\theta) = \sum_{l=0}^{\infty} A_l r^l P_l(\cos\theta)$$
(B.20)

Applying BC. 2 leads to the conclusion that all values of A_l are zeros apart from l = 1. This leads to the following expression for the temperature profile:

$$T_p = Ar \cos \theta \quad \text{for } 0 \le r \le a$$

$$T_f = Br \cos \theta + Cr^{-2} \cos \theta \quad \text{for } r \ge a$$
(B.21)

To find the values of the three constants, boundary conditions 2,3 and 4 are applied, which results in:

$$A = \frac{3k_f}{k_p + 2k_f}b$$

$$B = b$$

$$C = \frac{k_f - k_p}{k_p + 2k_f}b a^3$$
(B.22)

Substituting these constants in the temperature profile equations B.21, the final temperature profiles are:

$$T_p = \frac{3k_f}{k_p + 2k_f} b \ r \cos \theta$$

$$T_f = b \ r \cos \theta + \frac{k_f - k_p}{k_p + 2k_f} b \ a^3 \ r^{-2} \cos \theta$$
(B.23)

The temperature gradient across z axis can also be found:

$$\frac{dT_p}{dz} = \frac{3k_f}{k_p + 2k_f}b$$

$$\frac{dT_f}{dz} = \left(b + \frac{k_f - k_p}{k_p + 2k_f} \frac{-2 \ b \ a^3}{r^3}\right) \rightarrow \frac{dT_f}{dz} = b \ (for \ r \gg a)$$
(B.24)

Introducing effective thermal conductivity using spatial averages of the heat flux $\langle q_z \rangle$ and temperature gradient $\langle \nabla T_z \rangle$:

$$\langle q_z \rangle = -k_{eff} \langle \nabla T_z \rangle$$
$$\langle q_z \rangle = \frac{1}{V} \int_v q_z dv$$
(B.25)
$$\langle \nabla T_z \rangle = \frac{1}{V} \int_v (\nabla T_z) dv$$

where V is the volume across which the average is evaluated. The effective thermal conductivity can then be written as:

$$k_{eff} = -\frac{\int_{v} q_{z} dv}{\int_{v} (\nabla T_{z}) dv} = -\frac{\sum_{i=1}^{2} V_{i} q_{i,z}}{\sum_{i=1}^{2} V_{i} (\nabla T_{i,z})}$$
(B.26)

for i = 1,2, which represents the particles and the fluid region. Introducing volume ratio ϕ , the effective conductivity can be written in terms of particles and fluid respective volume contributions:

$$q_{i,z} = -k_i \frac{dT_i}{dz} \quad \text{and } V_i = V\phi_i \ (i = p, f)$$

$$k_{eff} = \frac{\phi_p k_p \frac{dT_p}{dz} + \phi_f k_f \frac{dT_f}{dz}}{\phi_p \frac{dT_p}{dz} + \phi_f \frac{dT_f}{dz}}$$
(B.27)

Substituting the temperature gradients B.24 and simplifying

$$k_{eff} = \frac{\phi_p k_p \frac{3k_f}{k_p + 2k_f} b + \phi_f k_f b}{\phi_p \frac{3k_f}{k_p + 2k_f} b + \phi_f}$$
(B.28)

Using the volume ratio of fluid in terms of particles volume ratio $\phi_f = (1 - \phi_p)$, and multiplying by $(k_p + 2k_f)$:

$$k_{eff} = \frac{3\phi_p k_p k_f + (1 - \phi_p) \ (k_p + 2k_f) k_f}{3\phi_p k_f + (1 - \phi_p) \ (k_p + 2k_f)}$$
(B.29)

Taking k_f as a common factor on the right side:

$$k_{eff} = \frac{k_f \left(k_p + 2k_f + 2\phi_p \left(k_p - k_f\right)\right)}{k_p + 2k_f - \phi_p \left(k_p - k_f\right)}$$
(B.30)

Finally, to present the enhancement effect of particles on the fluid thermal conductivity, the ratio of the effective thermal conductivity to the fluid thermal conductivity is written as follows:

$$\frac{k_{eff}}{k_f} = \frac{r+2+2\phi_p(r-1)}{r+2-\phi_p(r-1)} = 1 + \frac{3\phi_p(r-1)}{r+2-\phi_p(r-1)}$$
(B.31)

where $r = \frac{k_p}{k_f}$ is the ratio of particle conductivity to the fluid conductivity.

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