Droplet Behavior in Dense, Low Velocity Aerosols

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DROPLET BEHAVIOR IN DENSE, LOW VELOCITY AEROSOLS

By

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ABSTRACT
DROPLET BEHAVIOR IN DENSE, LOW VELOCITY AEROSOLS

Alexander F. Polley, B.S.
Marquette University, 2012

Rapid compression machines (RCM) are laboratory devices used to measure gas-phase fuel reactivity at conditions relevant to combustion engines. Test mixtures are generally prepared by rapidly compressing a gas phase fuel+oxidizer+diluent mixture to high pressure and temperature (e.g., 10-50 bar, 650-1000 K). It is extremely challenging to utilize diesel-relevant liquid fuels in these devices due to their involatility. One proposed method involves the delivery of an aerosol of suspended fuel droplets ($\approx 0.1$ mL$_{\text{fuel}}$/L$_{\text{gas}}$ at stoichiometric fuel loading) to the machine. The compression stroke of the RCM subsequently heats the gas phase of the aerosol thereby achieving vaporization of the fuel. The properties of the aerosol delivered to the RCM such as the droplet size distribution (DSD) are critical to ensuring successful execution of the experiments. For instance, the fuel droplets must be smaller than a critical threshold (e.g., $d_0 \approx 6 - 10 \mu$m) to ensure timely fuel evaporation and gas-phase mixing; in addition, the droplets must resist gravitational forces that could cause them to fall out of suspension. Low aerosol velocities are required in order to minimize fluid motion and thus heat loss from the compressed reacting gases during the RCM experiment. An aerosol model has been developed in this thesis project in order to understand the aerosol dynamics during the generation and machine delivery processes. Issues such as droplet impingement, coagulation, evaporation and settling can be investigated with the model, and thus the configuration (i.e., intake valve geometry, mixing chamber design, etc.) and operational characteristics (i.e., gas flow rates, fuel loading, etc.) of the aerosol RCM can be understood/improved/optimized. The system model is validated against gravimetric measurements using a mock up of a proposed delivery system. ‘Operating maps’ are generated for $n$-dodecane and $n$-hexadecane in oxygen + diluent mixtures covering a range of fuel loadings and delivery flow rates.
ACKNOWLEDGMENTS

Alexander F. Polley, B.S.

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<table>
<thead>
<tr>
<th>Symbol</th>
<th>Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>$A$</td>
<td>area</td>
</tr>
<tr>
<td>$A_v$</td>
<td>Avogadro’s number</td>
</tr>
<tr>
<td>$C$</td>
<td>Cunningham correction factor</td>
</tr>
<tr>
<td>$C_p$</td>
<td>heat capacity</td>
</tr>
<tr>
<td>$D$</td>
<td>diffusion coefficient</td>
</tr>
<tr>
<td>$D_n$</td>
<td>Dean number</td>
</tr>
<tr>
<td>$E$</td>
<td>molar growth rate of a particle (kmol/s)</td>
</tr>
<tr>
<td>$K$</td>
<td>coagulation coefficient (m$^3$/s)</td>
</tr>
<tr>
<td>$K_n$</td>
<td>Knudsen number</td>
</tr>
<tr>
<td>$L$</td>
<td>percent mass loss</td>
</tr>
<tr>
<td>$L_{char}$</td>
<td>characteristic length</td>
</tr>
<tr>
<td>$M$</td>
<td>molar mass</td>
</tr>
<tr>
<td>$N$</td>
<td>particle concentration (particles / kg$g$)</td>
</tr>
<tr>
<td>$P$</td>
<td>pressure</td>
</tr>
<tr>
<td>$Q$</td>
<td>volumetric flow, concentration of liquid molecules (kmol$\text{molecules}$/ kg$g$)</td>
</tr>
<tr>
<td>$R$</td>
<td>net concentration flux of liquid molecules (kmol$\text{molecules}$/ s$\cdot$kg$g$)</td>
</tr>
<tr>
<td>$R_o$</td>
<td>curve ratio</td>
</tr>
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<td>$Re$</td>
<td>Reynold’s number</td>
</tr>
<tr>
<td>$Sc$</td>
<td>Schmidt number</td>
</tr>
<tr>
<td>$St$</td>
<td>Stokes’s number</td>
</tr>
<tr>
<td>$T$</td>
<td>temperature (K)</td>
</tr>
<tr>
<td>$U$</td>
<td>velocity</td>
</tr>
<tr>
<td>$\bar{U}$</td>
<td>average velocity</td>
</tr>
<tr>
<td>$U_{TS}$</td>
<td>terminal settling velocity</td>
</tr>
<tr>
<td>$V$</td>
<td>volume</td>
</tr>
<tr>
<td>$X$</td>
<td>total number of DSD sectional bins</td>
</tr>
<tr>
<td>$d$</td>
<td>diameter</td>
</tr>
<tr>
<td>$g$</td>
<td>gravitational acceleration</td>
</tr>
<tr>
<td>$h$</td>
<td>enthalpy, height</td>
</tr>
<tr>
<td>$h_{conv}$</td>
<td>convection coefficient</td>
</tr>
<tr>
<td>$k$</td>
<td>thermal conductivity</td>
</tr>
<tr>
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</tr>
<tr>
<td>$m$</td>
<td>mass</td>
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<tr>
<td>$\dot{m}$</td>
<td>mass flow rate</td>
</tr>
<tr>
<td>$n$</td>
<td>moles</td>
</tr>
<tr>
<td>$n_j$</td>
<td>concentration of particles with size $n_j$ (kmol$\text{molecules}$/ kg$g$$\cdot$kmol$\text{particles}$)</td>
</tr>
<tr>
<td>$\dot{q}$</td>
<td>energy flux into the control volume</td>
</tr>
<tr>
<td>$r$</td>
<td>radius</td>
</tr>
<tr>
<td>$t$</td>
<td>time</td>
</tr>
<tr>
<td>$x$</td>
<td>distance</td>
</tr>
</tbody>
</table>

*Greek Symbols*
\[\Delta t\] \hspace{1cm} \text{time step}

\[
\chi \hspace{1cm} \text{fraction loss of particles}
\]

\[
\epsilon \hspace{1cm} \text{collision efficiency of two droplets}
\]

\[
\phi \hspace{1cm} \text{equivalence ratio}
\]

\[
\lambda \hspace{1cm} \text{mean free path}
\]

\[
\nu \hspace{1cm} \text{viscosity}
\]

\[
\theta, \phi \hspace{1cm} \text{angle}
\]

\[
\rho \hspace{1cm} \text{density}
\]

\[
\sigma \hspace{1cm} \text{diameter of a general particle}
\]

\[
\tau \hspace{1cm} \text{particle relaxation time, ignition delay time}
\]

\[
\omega \hspace{1cm} \text{acentric factor}
\]

**Subscripts**

- \(\infty\): far field
- \(1, 2\): general particles with 1 being larger than 2
- \(0\): \(t = 0\)
- \(B\): Brownian
- \(G\): gravitational
- \(LF\): laminar flow
- \(b\): tube bend
- \(c\): critical
- \(cs\): cross section of tube
- \(di\): disc
- \(dt\): delivery tube
- \(e\): exit
- \(evap\): evaporation
- \(g\): gas phase
- \(in\): inlet
- \(i, j, k\): DSD sectional bin numbers
- \(l\): liquid phase
- \(mc\): mixing chamber
- \(p\): particle
- \(rc\): reaction chamber
- \(sat\): saturation
- \(stem\): stem
- \(sys\): system
- \(tu\): straight tube
- \(v\): vapor phase
- \(vh\): valve housing
- \(w\): wall

**Superscripts**

- \(\ast\): dimensionless
Chapter 1

INTRODUCTION

This thesis describes the development and application of a model utilized to predict the behavior of dense, low velocity aerosols in a laboratory scale aerosol flow system. The primary purpose of the system is to deliver involatile, transportation-relevant fuels to a rapid compression machine (RCM). Experiments are performed with RCMs to understand the combustion kinetics of fuels. The data and chemical kinetic models developed from them have many important applications in the study and development of combustion engines. Aerosol fueling of RCMs and the associated ‘wet compression’ process has been proposed as a means of extending the utility of RCMs beyond gasoline-relevant components and surrogate blends to the regime of diesel type fuels.

The chapter begins with an introduction to the study of aerosol dynamics, especially concerning the modeling of aerosol transport, followed by a review of experimental equipment used to gather chemical kinetic data for engine-relevant fuels. Fuel loading of a few apparatuses is then discussed. The focus of this study is then presented and the chapter concludes with an overview of the thesis.

1.1 Study of Aerosols

An aerosol is defined as a solid or liquid that is suspended in a gas. Examples of liquid particle aerosols are clouds, fog, spray paint and medical inhalers. Examples of solid particle aerosols include smoke, smog, engine exhaust, and airborne dust. Aerosols can stay in suspension anywhere from a few seconds to a year depending on the physical properties of the particle and gas. Aerosol particles range from 0.001 to over 100 μm with
The General Dynamics Equation (GDE) is used to model the transport of aerosol particles within the gas phase and incorporates five mechanisms that lead to changes in the aerosol particle distribution:

\[
\frac{\partial n(d_p,t)}{\partial t} = C(d_p,t) + G(d_p,t) + N(d_p,t) + S(d_p,t) + R(d_p,t)
\] (1.1)

Here \(d_p\) is the particle diameter and \(t\) is time. Figure 1.1 presents sketches of these mechanisms where each is described next.

**Coagulation** \(C(d_p,t)\), occurs when two particles collide and become one. This happens as a result of particle motion caused by gravitational settling, velocity gradients, or electrostatic forces, as well as Brownian motion [2]. Brownian motion is the random motion of small particles in a gas due to the gas particles striking the suspended particles. This effect decreases as the suspended particles increase in size and are no longer affected by the gas particle momentum [1].
Growth, \(G(d_p, t)\), includes both condensation and evaporation of suspended particles. When the environment around a suspended particle becomes super-saturated (i.e. \(P_\infty > P_{\text{sat}}\)), the vapor phase will condense onto suspended particles and increase their size. The opposite is true when the environment is under-saturated. The suspended particles will decrease in size as some of the liquid phase evaporates [2].

Nucleation, \(N(d_p, t)\), will also happen when the gas phase of an aerosol is super-saturated. New suspended particles can be formed as the gas and liquid phases arrive at equilibrium.

Sources, \(S(d_p, t)\), account for all particle generation (e.g., from an atomizer) with the exception of nucleation.

Removal, \(R(d_p, t)\), occurs when particles fall out of suspension. This is a result of impaction, diffusion or gravitational settling onto surfaces. Diffusion is the dominant phenomenon causing removal for particles under 1 \(\mu\)m while impaction and settling have more influence for larger particles [1].

There are many fields that are concerned with the study of aerosol behavior. The remainder of this section will introduce aerosol behavior, physical properties of aerosols and major aerosol mechanisms studied in some of these fields. The section concludes with an overview of fuel aerosols, which are under investigation towards developing experimental equipment that is capable of acquiring chemical kinetic data for involatile, transport-relevant fuels.

Atmospheric Sciences

Aerosols in the atmosphere such as smog and clouds play a central role in the world’s climate system. These aerosols are a contributing factor in determining how much and what type of radiation reaches and leaves the earth [2]. Therefore, it is very important to have appropriate models that can facilitate the accurate prediction of climate behavior.

Primary aerosol dynamics are fairly well understood and the current focus in this field is on incorporating aerosol models efficiently into computational fluid dynamics (CFD)
The study of atmospheric aerosols includes the mechanisms of coagulation, growth, nucleation and removal. Models for coagulation, growth and nucleation are well studied and have been extensively verified. They are universal to studies where these mechanisms contribute to aerosol dynamics. Growth and nucleation can occur due to phase changes as well as chemical changes in the aerosol. Removal of aerosol particles occurs when rain drops fall and collect particles that are suspended in the air. This removal mechanism does not have much application outside of the field of atmospheric sciences.

The aerosols studied in atmospheric sciences are composed of dust, sea salt, water and various types of pollution. The aerosols are classified as smog (0.001 - 2 µm, <0.005 g/m³), clouds/fog (2 - 70 µm, 0.005 - 0.05 g/m³) or mist (70 - 200 µm, 0.01 - 0.1 g/m³) [1].

**Combustion Systems**

There is an ever increasing demand to reduce particulate emissions of combustion systems due to detrimental effects on the environment and human health. As a result, soot formation in flames is a major concern in combustion system design and has become a focus of many studies. One of the major components of kinetic models for soot formation is the soot particle dynamics.

The major mechanisms of particle dynamics involved in soot formation are nucleation, coagulation and growth. The products of gas phase reactions can nucleate as they become larger molecules with lower saturation pressures. These molecules will coagulate as they collide with each other and will also grow as a result of chemical reactions that occur at the surface of the droplet. Sub-models for the mechanisms in soot formation models have been taken largely from atmospheric aerosol studies [3].

Soot is a result of the incomplete oxidation of the carbon and hydrogen atoms originally contained in the fuel. Soot particles in diesel and gasoline engine exhaust range from 20 to 500 nm and are at concentrations of $10^5$ to $10^{10}$ particles/cm³ [4].
Drug Delivery

Certain medications are most effective when delivered directly to a person’s lungs where the medication can enter the bloodstream or be absorbed within a specific section of the lung. To achieve this, liquid solutions containing pharmaceutical compounds are atomized into small particles and delivered through a patient’s airway. During drug delivery, the aerosol passes through the mouth/nasal cavity, trachea, lung bifurcation, smaller branches and alveoli. The changes in direction along this path result in the deposition of larger aerosol particles along the early sections of the airway. Many studies have investigated the process in order to understand and predict how aerosol medications should be delivered.

Aerosol modeling in drug delivery focuses almost exclusively on removal mechanisms for aerosol particles. Early studies involved modeling and collecting data in simple setups using straight or bent tubes to model the airway. More recent studies simulate complex geometries with rough surfaces and use CFD packages to develop airway models that can very accurately predict deposition in human lungs. The methodologies used are applicable to all aerosols, however, the implementation can be very time consuming. Results from these studies have had little applicability outside of the field.

The composition of drug aerosols are also specific to the field. Aerosol particles are composed of active pharmaceutical ingredients dissolved in a combination of water, glycerin, propylene glycol or ethanol. The aerosol particles are commonly generated using ultrasonic, jet, micro-pump and vapor condensation nebulizers. These devices generate particles below $10 \mu \text{m}$ with concentrations on the scale of $10^4 - 10^8 \text{ particles/cm}^3$ before dilution with air [5].

Material Processing

Chemical vapor deposition (CVD) is a method of material processing which is used to coat a substrate with a thin film of material. The process starts with a precursor gas or gases being introduced inside of a chamber containing the substrate. The gases undergo a chemical reaction that produces a solid particle which is deposited on the substrate. For some CVD processes, this reaction occurs at the substrate surface [6]. In other processes,
solid particles form away from the substrate producing an aerosol inside the chamber. The particle size of the aerosol can be predicted to determine the impact on film quality [7].

Important mechanisms involved with CVD modeling are nucleation, growth and coagulation. In this field, nucleation and growth models involve a chemical change as the gas phase reacts to produce a solid that is deposited. Models developed and utilized in this field sometimes have applicability to combustion studies investigating particulate matter formation or engine exhaust behavior.

Some examples of materials that can be applied using CVD are silicon, tungsten, and silicon dioxide for semiconductors and indium gallium nitride (InGaN) for LEDs [6]. A study by Sharifi and Achenie [7] investigated the thin film production of zinc sulfide using CVD. The deposited particles were determined to have an average diameter of 2.35 nm with a concentration of roughly $7 \times 10^{11}$ particles/cm$^3$. This is a very large particle density compared with other fields, but the overall mass density is much lower due to the small particle size.

**Aerosol Sampling**

Aerosols are present in many systems and often need to be monitored for a variety of reasons including safety, regulation compliance and process design. Ideally, this data would be collected without interfering with the aerosol, but this is usually not possible. At nuclear power plants and nuclear waste disposal facilities the radioactivity of aerosols needs to be monitored in accordance with Environmental Protection Agency (EPA) regulations. As a result, aerosol samples are collected in one location and analyzed at another. This requires transport of the aerosol through a series of pipes which results in some aerosol particle loss. In order to infer the actual aerosol properties from the sampled and measured ones, the effect of transport system deposition needs to be understood [8].

Similar to drug delivery, aerosol sampling models have focused primarily on particle removal. Models for pipes and bends are created using results from CFD calculations that investigate a range of flow conditions. While these models are primarily applicable to specific geometries, they are often non-dimensionalized and thus can have a wide range of applicability to engineered systems where suspended particles have various
size, composition and density.

**Indoor Air Quality**

Since the majority of the work force spends significant amounts time inside of buildings, effects of indoor air quality is becoming a big concern. Studies indicate that the quality of the air has a direct effect on the health of building occupants [9]. The chemical compositions as well as the physical properties of particles suspended in the air both contribute to the overall impact. Specifically, ultrafine particles (i.e. < 100 nm) with low-solubility have been shown to have adverse effects on a person’s health. Aerosols generated within a building can be different sizes and be generated in multiple areas. These aerosols interact and are eventually inhaled. It is important to know the properties of the inhaled aerosol and if building modifications can have any influence [10].

Air quality models incorporate all general mechanisms of the GDE. Numerical studies often use common coagulation, growth and nucleation models and even incorporate sources for aerosols generated within buildings. Removal models include losses to ventilation ducts as well as to room objects, walls and floors. Studies can investigate building-scale to room-scale scenarios depending on the objectives.

Aerosol particles that have been studied are composed of solids such as polystyrene latex, titanium dioxide, aluminum, polytetrafluoroethylene (PTFE), and granite. Particle sizes range on the order of 1 nm to 10 \( \mu \text{m} \) while concentrations have a maximum of \( 10^6 \) particles/cm\(^3\) [10].

**Involatile Fuels**

Understanding the reaction kinetics of a fuel during combustion is very important towards the development of modern combustion systems that can meet efficiency and emission targets. In order to collect data and develop models for the combustion chemistry, fuels must be reacted under tightly controlled conditions. One requirement for such tests is that the fuel must be fully vaporized prior to the onset of chemical reaction processes. This poses a problem for fuels with extremely low vapor pressures such as those representative of diesel. One method that has been proposed to overcome this problem is
to prepare the liquid phase fuels as an aerosol which can be evaporated inside test
equipment due to ‘wet compression’ (i.e., the volumetric compression heating of the gas
phase) before combustion begins.

Loading a fuel aerosol with an appropriate droplet size distribution (DSD) and
overall fuel concentration is a major challenge. In order for the fuel to completely
evaporate and mix with the oxidizer and diluent gas, droplets must be smaller than a
specific size based on the fuel and equipment [11, 12, 13]. In order to achieve this loading
specification, the effect of the aerosol generation and delivery systems on the DSD must
be known.

The model developed in this study is used to predict changes in DSD along an
aerosol delivery system. The model can also track fuel fall out as well as the DSD and
equivalence ratio ($\phi$) that remains suspended in the reaction chamber. It incorporates
sub-models for coagulation, growth and removal and draws upon findings in the fields
mentioned in this section. Sub-models for coagulation and growth are applicable, and
removal sub-models come from work investigating geometries that are similar to the
delivery system for a representative experimental apparatus which is discussed in the next
section.

In order to study engine-relevant combustion chemistry, the gas phase of an
aerosol will contain varying levels of oxygen concentration ranging from 1 - 21% O$_2$. This
means that an aerosol with a stoichiometric ratio of dodecane plus air, for example, needs
to have a concentration on the order of $10^6$ particles/cm$^3$ at standard temperature and
pressure when the droplet diameter is 4 $\mu$m [12]. Smaller droplet sizes will require higher
concentrations. The properties of the fuel and the conditions to be studied dictate the
density and maximum allowable droplet size of a fuel aerosol.

1.2 Engine-Relevant Chemical Kinetics

Research focusing on the combustion chemistry and the underlying fuel/oxidizer/diluent
kinetics has led to major reductions in emissions of harmful compounds (e.g., NOx, SOx)
and particulate matter in combustion systems over the past 50 years. These efforts involve
developing an understanding of chemical pathways by proposing reaction models (i.e. kinetic mechanisms) and testing the validity of these models against fundamental experimental data as well as data from operating engines. Work is ongoing to refine models and to create new models that can account for more complex chemistries.

The major fuels in transportation-relevant combustion systems are derived from petroleum (e.g., gasoline, diesel and jet fuel) and combustion generally occurs with peak temperatures ranging from 1400 - 2800 K. As combustion systems evolve, systems will utilize fuels with different molecular structures (e.g., butanol and biodiesel) and will operate utilizing new combustion regimes (e.g., HCCI, DCDI, etc.) at temperatures lower than traditional systems. These new systems will require continued work towards developing an understanding of the combustion process in order to design systems that meet performance and emission targets.

The remainder of this section will introduce some experimental equipment that is used in fundamental studies of the chemical kinetics of fuels undergoing combustion. Unless otherwise cited, the overview is a summary of work by Griffith & Mohamed [14].

1.2.1 Flow Reactors

Flow reactors can be classified as laminar flow (LFR), turbulent flow (TFR) or well stirred (WSR) reactors. Examples of equipment setups can be seen in Figure 1.2. In these
systems fuel, oxidizer and diluent gas are premixed and preheated before entering the reactor. In the case of the LFRs and TFRs, the test mixtures flow continuously through a cylindrical configuration and measurements are made at different locations along the reactor length. In the WSR, the test mixtures are introduced into a spherical vessel and are mixed by either the test mixture jet or a mechanical stirrer with measurements made at the reactor exit. Residence times are controlled by modifying the flow rate of the test mixture.

Flow reactors are generally operated at pressures from 0.1 - 1 MPa and equivalence ratios from 0.25 - 4.0. Temperatures in flow systems are well controlled in order to isolate the effect of temperature on chemical kinetics. The temperature range of experiments generally range from 600 - 1200 K and can either be held constant or, in the case of LFRs and TFRs, can be gradually increased as the test mixture proceeds along a tube. Test mixtures need to be dilute (e.g., 1% O\textsubscript{2}) in order to minimize the heat released during reaction so that chemically-induced thermal gradients do not exist in the test mixture. Generally the concentration of oxygen in the mixtures must be kept below three percent by mass. The residence time of the test mixture ranges from 10 to 3000 ms.

In flow reactors, the concentrations of reaction intermediates are recorded during an experiment. Concentrations can be measured by either direct sampling of the test mixture or by spectroscopic methods. When using direct sampling, the probe must be designed to not interfere with the reaction and must ensure that reactions are stopped after the gas enters the probe. After sampling, the species are delivered to analytical equipment, such as a mass spectrometer, where the concentrations are measured. Spectroscopic methods, such as laser induced fluorescence, have the benefit of not interfering with a test mixture during measurement.

An example of experimental data for \textit{n}-hexadecane oxidation in a WSR can be seen in Figure 1.3 [17]. Measurements of mole concentrations for different chemical species are plotted against the reactor temperature. These results can be used to validate kinetic models for the oxidation of the fuel.
1.2.2 Shock Tubes

Shock tubes (ST) use a rapidly traveling pressure wave to compression heat a test mixture to high temperature and pressure. The pressure wave can be generated through a number of ways including via the fracturing of a diaphragm separating the test mixture from a higher pressure driver gas. A diagram of a typical shock tube can be seen in Figure 1.4. A vacuum pump is used to evacuate the low pressure section of the tube followed by the loading of the test mixture. Optical access at the end wall of the shock tube allows for measurements to be taken. Any direct sampling equipment would also be located near the end wall (e.g., see ref [19]).

After the diaphragm bursts during an ST experiment, an initial shock wave travels
down the tube ahead of the driver gas, heating and raising the pressure of the contents of the test section. The shock wave is reflected off the end wall and further raises the temperature and pressure of the test gas. Generally, the test mixture is held at the elevated temperature and pressure for 1 to 2 ms before experimental conditions deteriorate as the driver gas reaches the end wall. Pressures and temperatures after the reflected shock passes through the test section range from 0.1 to 10 MPa and 900 to 2500 K, respectively. Similar to flow systems, the concentration of fuel needs to be kept low in order to control the homogeneity of the test gas. Equivalence ratios of ST experiments generally vary from 0.25 to 2.0. Higher equivalence ratios can lead to soot deposits on the reaction chamber walls which generally must be cleaned between tests.

Measurements of the concentration of species in ST experiments are collected in the same manner as flow systems. The measurements are taken at the end wall of the ST where the reaction conditions remain stable the longest. The pressure is also measured along the test section to follow the progress of the shock wave and determine the shock strength. Pressure measurements near the end wall are used to characterize the auto-ignition delay time of a fuel. An example of experimental results from a study of \textit{n}-decane can be seen in Figure 1.5 [20]. The plot shows both the pressure and OH*
concentration as a function of time. In the sidewall pressure trace, the first jump in pressure represents the passing of the initial shock wave. The next jump in pressure represents the reflected shock wave, while the final increase in pressure signifies autoignition of $n$-decane. The OH* concentration trace shows one large increase which also due to autoignition of $n$-decane. The pressure and OH* trace are used to determine the ignition delay time, $\tau$, at the specified condition. The ignition delay time is a cumulative measure of the fuel mixture reactivity and is often measured during studies because it is not as complicated as measuring species concentrations.

### 1.2.3 Rapid Compression Machines

The reaction section of a Rapid Compression Machine (RCM) is similar to the piston and cylinder configuration that is present in IC engines. The piston in an RCM is designed to travel at speeds between 5 and 15 m/s and locks into position at the end of the compression stroke. A diagram of an RCM is presented in Figure 1.6. The reaction chamber is typically loaded using the same method as described for the ST. To initiate an experiment, the driver piston is locked in place (e.g., via hydraulic fluid) while high pressure is built up behind it. The driver piston is then released causing the reaction piston to compress the contents of the reactor chamber. A hydraulic locking chamber, or something similar, is used to hold the reaction piston at the peak of the compression stroke, providing a constant volume for the duration of the experiment.
Typical compression ratios in an RCM range from 10 to 15:1. The pressure and temperature achieved by the test mixture after compression can cover 0.5 to 5 MPa and 500 to 1100 K, respectively. RCMs are capable of sustaining significant heat release during the experiment due to their design operation and therefore test mixtures can contain 5 to 21% O\textsubscript{2} with equivalence ratios from 0.25 to 2.0. However, RCMs must also be designed to control the temperature and uniformity of the test mixture before a reaction initiates. Another distinction of the RCM is that measurement times can be as long as $\approx$150 ms.

Data collection in RCMs is similar to that in STs with sampling probes and optical access located near the end wall. An example of results from experiments using $n$-decane and air mixtures can be seen in Figure 1.7 [22]. The plot shows the pressure trace near the end wall of the RCM for five different compressed mixture temperatures. At the experiment conditions, $n$-decane has a two-stage ignition as shown by the two pressure increases after then end of compression. The ignition delay times can be calculated based on the pressure traces.

### 1.2.4 Equipment Comparison

Various types of experimental equipment are needed to cover the entire range of conditions that are of interest in the study of engines. While there is overlap between the equipment, each apparatus also has conditions where it is the only effective test method.
Figure 1.8: Ignition delay time as a function of inverse temperature from RCM (●, ○) and ST (▲, △, +) studies of n-decane/air mixtures at compressed pressures of 13-14 bar [22].

For instance, flow systems can investigate fuel reacting over large timescales. STs can measure fuel behavior at high temperature and pressure. RCMs are useful for low temperature combustion at oxygen concentrations that are equivalent to the atmosphere.

Data from many experiments is collected and can be plotted to gain a complete understanding of fuel behavior. An example of a plot of ignition delay of n-decane as a function of inverse temperature created using data from various studies can be seen in Figure 1.8. The RCM results from Kumar [22] are compared with ST results from other studies. The plot shows that the RCM was able to investigate lower temperature regions than ST studies at similar pressures. This is important to gain a full understanding of fuel’s decomposition and oxidation behavior.

1.3 Fuel Loading in STs and RCMs

The fuel loading of STs and RCMs was briefly discussed in the previous section. This section will expand on the discussion by introducing the methods of partial pressure and aerosol fuel loading of the experimental equipment.
1.3.1 Partial Pressure Method

Fuel loading into single shot equipment investigating gas phase chemistry has historically been achieved by using the method of partial pressures. In this method, a mixing tank is first evacuated and then the fuel is added. The fuel can be added by direct injection of a desired amount of fuel, which will then evaporate inside the mixing tank, or the fuel can be loaded as a vapor and the amount can be controlled by monitoring the pressure of the tank. Once the desired amount of fuel is loaded, the diluent gas and oxidizer are added to the tank sequentially. The mixture may be mechanically mixed first and can be stirred for 3 hours or more to ensure homogeneity.

The reaction chamber of the experimental apparatus is first evacuated and then opened to the mixing chamber. The gas mixture fills the reaction chamber as a results of the pressure difference and is then isolated from the mixing chamber. The experiment can then be initiated after the gas is in thermal equilibrium with the walls of the reaction chamber.

The partial pressure method is useful for fuels that have a sufficient vapor pressure to provide the desired amount of fuel for an experiment. If the fuel has too low a vapor pressure, the temperature of the mixing tank and experiment apparatus may be increased in order to increase the vapor pressure of the fuel. This method will work for some fuels, but must be done carefully for the following reasons.

For a single component fuel the first issue of concern is to ensure that there are no cool locations within the apparatus to which the higher temperature vapor may be exposed. If these are present, the fuel may condense and ‘contaminate’ experimental results. The other area of concern is the stability of the fuel. At elevated temperatures the fuel may begin to decompose and change chemically. This too will cause any experimental measurements to be invalid.

For multi-component fuels, an experimenter needs to be concerned with distillation of the fuel. To ensure this does not happen, all fuel needs to be vaporized. If any is not, the portions with higher boiling temperatures will stay in the liquid phase and the gas phase will have a different composition than the fuel that is to be tested. Cool locations
will also result in the less volatile components to condense first resulting in liquid ‘contamination’ and a gas phase with a different composition.

1.3.2 Aerosol Method

One method proposed to overcome the problems associated with the partial pressure method for high boiling point fuels is aerosol loading. In this method, the fuel is introduced into the test section of the ST or RCM as small suspended liquid droplets. The droplets are evaporated in the test equipment during the initial compression heating process before chemical reactions take place.

In a shock tube, evaporation occurs between the arrival of the initial shock wave and arrival of the reflected shock wave. The initial compression initiates evaporation by increasing the temperature of the gas and thus liquid phase and by breaking up droplets into smaller fragments, thereby increasing the surface area of the liquid phase. Facilities are currently in use at Stanford University [23, 24] and Texas A&M University [25] where aerosol shock tubes are utilized.

A schematic of the initial aerosol ST developed at Stanford University is presented in Figure 1.9. The top image shows the aerosol as it is loaded into the ST. The bottom image shows how the aerosol is vaporized as the initial shock wave moves toward the end wall of the ST. This system was designed to employ ultrasonic nebulizers (Ocean Mist®)
Figure 1.10: A diagram showing the second generation aerosol ST at Stanford University. The aerosol is mixed in a large tank and introduced through a gate valve in the ST end wall [27].

[26] to generate droplets that are carried along the delivery tube by the gas phase oxidizer and diluent and into the endwall of the ST. The aerosol is delivered to the ST test chamber through multiple poppet vales that are closed after loading to seal the test chamber. With this arrangement, the larger droplets are expected to fall out of suspension during the delivery leaving only droplets smaller than 20 \( \mu m \) suspended in the aerosol [26].

Stanford’s second generation aerosol ST, shown in Figure 1.10, utilizes a 22 L mixing tank with a plenum to mix and suspend the droplets in the test gas. Better spacial uniformity of the aerosol was a primary motivation for the modified design. The aerosol is generated through the use of ultrasonic nebulizers in the mixing tank and then the sliding valve at the endwall of the ST is opened. A valve to the dump tank is opened and the aerosol flows through the test chamber due to the pressure difference between the ST and the dump tank. Once the aerosol is loaded, the valve at the endwall and the valve to the dump tank are closed. A second sliding valve connecting the driver and driven section is then opened and the experiment is initiated.

The aerosol ST at Texas A&M University operates in a fashion similar to the first generation Stanford aerosol ST and can be seen in Figure 1.11. It, however, utilizes an AGF 2.0 Aerosol Generator (Palas Technology) to provide an aerosol with droplets smaller than 0.8 \( \mu m \). The aerosol is introduced to the ST through a delivery tube far away from the endwall. A small opening at the endwall is connected to a pump that provides
1.4 Aerosol Loading in an RCM

The development of an aerosol system that could be coupled to an RCM is a focus in the Combustion Lab at Marquette University. A preliminary generation and delivery system was designed and tested and will be discussed in the following sections. The inadequate performance of this system was a motivation for the current work.

1.4.1 Configuration

A schematic of the preliminary aerosol system can be seen in Figure 1.12. The system is composed of a mixing chamber, delivery tube, valve housing, reaction chamber and an exit manifold. Droplets are generated at the top of the mixing chamber through a medical grade ultrasonic mesh-type nebulizer and are diluted with gas to the desired concentration. The aerosol then travels through the other components of the system before exiting.

One important behavior of the aerosol that must be considered during a particular...
Figure 1.12: Diagram of the aerosol RCM mock up at Marquette University

test is the fall out of fuel droplets along the delivery system. This is important because any fall out alters the fuel to air ratio that is delivered to the reaction chamber, which is a key parameter that must be controlled in RCM experiments. It is also important to characterize the fall out within the reaction chamber because wall contamination of liquid fuel could affect the RCM experiment results.

1.4.2 Initial Aerosol Loss Results

Experiments were carried out to measure the aerosol loss for various delivery system arrangements. In the initial system design, close to 10 % of the aerosol mass was lost in the components the make up the reaction chamber (i.e. the valve housing, reaction cylinder and exit manifold). This extent of contamination in an RCM would not be acceptable for an experiment. Redesign of the valve housing reduced the fallout in the reaction chamber components to ≈ 5 %, but this is still substantial.

Other factors influencing aerosol droplet losses are constriction and interference of the aerosol flow in the valve housing. For example, the percent lost in the aerosol system increases by ≈ 2.0 % when a hose barb is used to attach the delivery tube to the valve housing where the flow area is reduced by 42 %; losses further increase ≈ 4 % when a poppet valve is added.

Minor changes made to the delivery system configuration to reduce flow restriction have a substantial effect on performance, however, changes to the DSD of the aerosol is unknown because adequate measurement devices are not available in the lab. Because of
these features it was determined that a fundamental understanding of aerosol behavior within the delivery system is needed where effects of system configurations and operational parameters (e.g., fuel loading, flow rate, etc.) can be assessed. A model which is capable of capturing the droplet dynamics could be used towards this, with the simulation results employed towards improving the system design. The model could also be used to explore system and operating conditions that might be difficult to test outside of an operational aerosol RCM.

1.5 Overview of Computer Model

A wide variety of aerosol models presented in the literature have been assembled for specific applications. None is quite applicable towards the goals of this study; though drug delivery models are close, they are cumbersome to utilize. Atmospheric models provide a good base from which to start as the coagulation and growth components of these models are universal to all aerosols. Removal mechanisms, on the other hand, are very specific to the problem or configuration being studied. Aerosol models developed by Anand, et al. [8] and von der Weiden, et al. [30], for instance, incorporate removal mechanisms as an aerosol flows through sampling systems composed of tubes. However, these models do not include the effects of coagulation and evaporation. These two factors are expected to play a large role in the delivery system where the temperature is elevated and the particle concentration is high.

In order to study and eventually design the delivery system for an aerosol RCM, a new model is developed here that integrates existing sub-models capable of accounting for the physical phenomenon that occur within the fuel delivery system.

1.6 Outline of Thesis

The outline of this thesis follows. In Chapter 2 the description of droplet populations and theory pertaining to droplet motion will first be discussed. The theory behind modeling the aerosol dynamics in a fuel delivery system is next presented. Discussion focuses on the development of applicable mechanisms and algorithms.
Chapter 3 reviews the methodology of the model study. Specific equations and how they are utilized within the aerosol model are discussed.

Chapter 4 presents model results and compares these with experimental measurements to demonstrate validity. The procedures of the experiments to measure the aerosol loses in the preliminary delivery system are presented. Additional simulation results are presented which explore effects of various operational parameters on the system performance. Behavior is investigated covering a range of temperatures, pressures, aerosol generators and bath gases, as well as fuel loading and flow rates. Modifications that could improve the system and model performance are suggested.

The report concludes with a summary in Chapter 5.
Chapter 2

LITERATURE SURVEY & THEORY

To create a model which adequately describes how the fuel DSD evolves within the aerosol fueling system the primary mechanisms affecting single droplets and droplet pairs must be taken into account. These mechanisms, which were introduced in Chapter 1, have been studied in depth in the literature. Of the five mechanisms reviewed, only three are important in the fuel delivery system and need to be included in the model: coagulation, evaporation and removal. Sources and nucleation are not included in the current model. In modeling the system, the aerosol generated by a nebulizer is specified as an initial input and there are no other sources in the system. Therefore, it is not necessary to include a source mechanism.

The mechanism for nucleation is not included because it is expected that the vapor phase of the fuel will not become over-saturated so that this mechanism will not have a significant effect. Over-saturation of the vapor phase in the system would only be expected if the initial bath gas were over-saturated, or there was a substantial drop in temperature within the system after the vapor phase becomes saturated. Both of these conditions are not expected in the operation of the fuel delivery system.

This chapter begins with a discussion of how aerosol populations are handled in a model followed by a review of droplet motion. Next, relevant mechanisms are presented and the chapter concludes with a presentation of the methodology used to incorporate the mechanisms into a system model that is capable of tracking the evolution of a droplet size distribution.
Figure 2.1: The same DSD shown as (a) particle and (b) volume fraction distributions.

2.1 Droplet Populations

In the study of aerosol dynamics, the most important factors are the size and quantity of the droplets present. The size of the droplets can be identified by molecules, mass, volume or diameter with concentrations expressed per m$^3$ or per kg of gas. These two factors are represented by the droplet size distribution (DSD). Two representations of a DSD can be seen in Figure 2.1.

Figure 2.1a shows the particle fraction distribution as a function of droplet diameter, while Figure 2.1b represents the volume fraction distribution of the same aerosol. The difference in appearance is due to the fact that the volume of a particle is related to its diameter cubed. For example, one 10 $\mu$m droplet has a volume equal to one thousand 1 $\mu$m droplets.

Because of the various ways to represent an aerosol population, averaging of the particle diameter for droplet populations often presents some confusion. An aerosol distribution can be represented by count, length, area or volume as a function of droplet diameter and can be averaged by these four properties as well. This results in 16 different methods of specifying the average particle diameter.

In addition, the average may be taken as the Stokes or aerodynamic diameter. The Stoke’s diameter is defined as the diameter of the sphere that has the same density and settling velocity as the aerosol particle. Whereas the aerodynamic diameter is defined as the diameter of the unit density sphere ($\rho_p = 1$ g/cm$^3$) that has the same settling velocity of the particle [1].
The method used in a particular model or analysis depends on the type of study and what parameter is important. For example, when studying the health effects of an aerosol, the count average is often used because the number of inhaled particles is important. On the other hand, for the present study of fuel aerosols, the volume average is more applicable because the need to vaporize all of the particles is important, in particular the largest ones which require the greatest evaporation time during the piston compression process.

When studying aerosols it is critical to understand how droplet populations are presented. See Chapter 4 in Hinds [1] for a complete discussion of particle size distributions and statistics.

2.2 Droplet Motion

One important area to cover before discussing mechanisms affecting the DSD of an aerosol population is droplet motion in aerosols. This plays a major role in the removal mechanisms that will be presented later. This section is a highlight of important information regarding particle motion as presented by Hinds [1].

As an object moves through a gas, a drag force is generated that resists the motion of the object. This phenomenon was first investigated by Newton when studying the motion of cannonballs in air. The particle Reynolds number of a cannonball is about 1000, and at this condition, viscous forces of the air are negligible compared to the inertial forces of the cannonball.

Aerosol particles, on the other hand, are generally very small and move slowly. They exist in conditions where the particle Reynolds number, \( \text{Re}_p \) is less than 1 and inertial forces of the particle are negligible compared to the viscous forces of the air. This region is referred to as the Stokes region. Solving the Navier-Stokes equations under these conditions results in Stokes law where the drag force on a particle can be described as

\[
F_D = \frac{\pi \rho_g d_p^2 U_p^2 C_D}{8}
\]

(2.1)

where \( C_D \) is the drag coefficient, \( \rho_g \) is the density the gas and \( U_p \) is the relative particle
velocity. The drag coefficient for spherical particles is equal to $24/Re_p$ in the Stokes region.

Setting the drag force equal to the force of gravity and solving for velocity determines the terminal settling velocity of a particle

$$U_{TS} = \frac{\rho_p d_p^2 g}{18\mu_g}$$  \hspace{1cm} (2.2)$$

where $g$ is gravitational acceleration and $\mu_g$ is the dynamic viscosity of the gas.

The equation needs to be corrected for particles that are smaller than 1 $\mu$m because the assumption that the relative velocity of the gas at the particle surface is zero does not hold. This can be done by using the Cunningham correction factor which reduces the drag force and increases the settling velocity by

$$F_D = \frac{\pi \rho_g d_p^2 U_p^2 C_D}{8C}, \quad U_{TS} = \frac{\rho_p d_p^2 g C}{18\mu_g}$$  \hspace{1cm} (2.3)$$

An expression of the Cunningham correction factor in various gases [31] is

$$C = \begin{cases} 
1 + \alpha Kn, & \text{if } Kn < 0.4 \\
1 + [\alpha + (1.647 - \alpha)e^{-\gamma/Kn}] Kn, & \text{if } 0.4 \leq Kn \leq 20 \\
1 + 1.647 Kn, & \text{if } Kn > 20
\end{cases}$$  \hspace{1cm} (2.4)$$

The values for $\alpha$ and $\gamma$ are determined from fits to experimental data. In the current work air and $N_2$ have $\alpha$ and $\gamma$ values of 0.78 and 1.207, respectively, while Ar has values of 0.85 and 1.227. The Knudsen number is

$$Kn = \frac{2\lambda_g}{d_p}$$  \hspace{1cm} (2.5)$$

with the mean free path of the gas calculated by

$$\lambda_g = \frac{\mu_g}{0.491\rho_g c}$$  \hspace{1cm} (2.6)$$
and the mean velocity of the gas molecules defined as

\[ \bar{c} = \sqrt{\frac{8R_uT}{\pi M_g}} \]  

(2.7)

The relaxation time of a particle is the time it takes for a particle to adjust its velocity to new conditions. It is commonly used in conjunction with the terminal settling velocity. It is generally assumed that an aerosol particle is traveling at its terminal settling velocity since the relaxation time of a 1 µm particle is on the order of $10^{-6}$ s. In the Stokes region the relaxation time is defined as

\[ \tau = \frac{\rho_p d_p^2 C}{18 \mu_g} \]  

(2.8)

The stopping distance of a particle is then defined as

\[ S = U_p \tau \]  

(2.9)

This is the distance that a particle traveling at velocity $U_p$ will need in order to stop and is an important factor in describing the curvilinear motion of aerosol particles.

Curvilinear motion is the motion of a particle on a curved path. It is characterized by the Stokes number, $St$, which is the ratio of the stopping distance to a characteristic length, $S/L_{char}$. The significance of the Stokes number is that it represents how closely particles will follow the stream lines in curvilinear flow. For example, when stream lines curve to avoid an obstacle, particles with high Stokes numbers are more likely to collide with the obstacle while particles with lower Stokes number will most likely follow the stream lines and avoid the obstacle.

The motion of particles determines how they interact in an aerosol. These interactions will be the focus of the following section.
2.3 Mechanisms in Aerosol Dynamics Models

The mechanisms that result in changes to an aerosol population have been studied and refined for many years. Coagulation and evaporation models apply generally to most aerosol systems, whereas removal models are specific to the geometries that are encountered by an aerosol. This section introduces the theory behind these mechanisms.

2.3.1 Coagulation

As described earlier, coagulation occurs when two particles come into contact and join to form one larger particle. This affects a DSD by decreasing the droplet concentration (particles/m$^3$) and increasing the droplet size. The equation for coagulation is

$$ C(d_p,t) = \frac{1}{2} \int_0^{d_p} K(d_p - \sigma, \sigma) n(d_p - \sigma, t) n(\sigma, t) \, d\sigma - n(d_p, t) \int_0^\infty K(\sigma, d_p) n(\sigma, t) \, d\sigma \quad (2.10) $$

where $K$ is the sum kernel for the coagulation coefficient, $n(d_p, t)$ is the particle distribution function and $\sigma$ is the diameter of a general particle. The first term on the right hand side of the equation represents particles that are size $d_p$ after coagulation, while the second term represents coagulation of particles that are size $d_p$ with other particles.

The physical phenomena that cause aerosol particles to combine include Brownian motion, gravitational settling, turbulent inertia, as well as laminar and turbulent shear. Brownian motion may also be referred to as thermal coagulation, while the other mechanisms are referred to as kinematic coagulation. Other forces that affect the coagulation of aerosol particles include inter-particle, hydrostatic, thermal and electrostatic [1, 2, 32]. Each of the physical processes will be discussed in the following section, while discussions of the forces can be found in the cited references.

As mentioned earlier, Brownian motion (i.e. particle diffusion) is the random motion of solid or liquid particles caused by collisions with gas molecules. When particles are small, the force of a striking gas molecule is enough to alter the trajectory of suspended particles. As particles increase in size, the effect of the striking gas molecule
diminishes. The particle diffusion coefficient is defined as

\[ D_p = \frac{k_b T_g C}{3\pi \mu_g d_p} \]  

(2.11)

where \( k_b \) is Boltzmann’s constant and \( T_g \) is the gas temperature.

When multiple particles are in an enclosed space, particle diffusion will cause some of the particles to collide with each other. Collisions occur when the distance between the center of the particles is equal to or less than the sum of the particles radii. Brownian coagulation is the major coagulation mechanism for particles smaller than 1 \( \mu \text{m} \), but still contributes to overall coagulation rates above 1 \( \mu \text{m} \) [32]. The equation for the Brownian coagulation coefficient between two particles is

\[ K_B(r_1, r_2) = 4\pi (D_{p,1} + D_{p,2}) (r_1 + r_2) \]  

(2.12)

Gravitational coagulation occurs due to the difference in terminal settling velocity of different sized drops. Larger drops fall faster than smaller ones and these will combine as they come into contact. This mechanism can be neglected for particles below 1 \( \mu \text{m} \) but becomes important as particles become larger [2]. The equation for the gravitational coagulation coefficient is

\[ K_G(r_1, r_2) = \epsilon \pi (r_1 + r_2)^2 (U_{TS,1} - U_{TS,2}) \]  

(2.13)

where particle 2 has the a smaller radius and \( \epsilon \) is the collision efficiency of the droplets which depends on the particle diameters.

A similar form of coagulation occurs in laminar flow. Adjacent particles in the flow move at different speeds. Faster particles are brought into contact with the slower particles and combine when they contact. This generally only effects particles below 1 \( \mu \text{m} \) which are able to follow stream lines. The equation for the coagulation coefficient due to laminar shear is

\[ K_{LF}(r_1, r_2) = \frac{4}{3}\Gamma(r_1 + r_2)^3 \]  

(2.14)
Figure 2.2: Brownian and gravitational coagulation rates of a 1 µm particle with particles ranging from 0.1 - 10 µm [32].

where $\Gamma$ is the velocity gradient perpendicular to the direction of flow. Laminar coagulation is usually not significant because large velocity gradients (e.g., 60 s$^{-1}$) are necessary in order to be of the same magnitude as Brownian coagulation [2].

Particles can also be brought into contact with one another in turbulent flow, however this phenomenon is much more complex and is only partially understood [32]. Turbulent coagulation can be segregated into shear and inertial components. A discussion of the kernels can be found in the literature but will not be presented here because the proposed delivery system and model to be used for it are expected to operate within the laminar flow regime.

A comparison of the Brownian and gravitational coagulation mechanisms for a 1 µm particle interacting with various sized particles is presented in Figure 2.2. The plot illustrates that for particles smaller than about 1 µm, Brownian motion is the dominant force. However, as the particle size increases, gravitational coagulation becomes an important factor.
2.3.2 Evaporation

The evaporation of aerosol droplets is controlled by the rate of diffusion of the vapor throughout the surrounding gas \([2, 33]\). Often, the rate of heat diffusion to the droplet to supply the vaporization enthalpy, as discussed in the next chapter, is assumed to be infinite, and thus ignored in many models. In the continuum regime, the steady flow of vapor from the surface of a droplet is defined by Maxwell’s equation:

\[
G(d_p, t) = -4\pi r_p D_v (\rho_{v, sat} - \rho_{v, \infty})
\]  

(2.15)

where \(m_p\) is the particle mass, \(D_v\) is the vapor diffusion coefficient into the bath gas, \(\rho_{v, sat}\) is the assumed vapor concentration at the droplet surface and \(\rho_{v, \infty}\) is the vapor concentration at the far field.

This equation is not valid as the particle diameter approaches the mean free path of the diffusing vapor \([2]\). Evaporation then lies in the transition regime and a correction factor can be applied to Maxwell’s equation. Various correction factors have been proposed by multiple investigators though all approaches provide similar results. One commonly used factor was proposed by Fuchs and Sutugin \([34]\) and is presented in Chapter 3.

The theory discussed above has been developed for droplets with little relative motion compared to the gas phase. This ideal case is not applicable for larger aerosol droplets (e.g., \(\approx 5 \mu m\)) where the settling velocity introduces relative motion between the droplet and gas phase. In this case, evaporation is enhanced by the gas motion past the droplet and Davies \([33]\) proposed another correction to Maxwell’s equation to account for this. Davies’ formulation is also presented in Chapter 3.

2.3.3 Removal

An aerosol in the fuel delivery system under investigation will be transported through a mixing vessel where the fuel droplets are entrained with the oxidizer and diluent gas, circular tubes that can be vertical, horizontal or curved which transport the aerosol from the mixing chamber to the reactor as well as the cylinder and disc geometries of a
poppet-type valve. All of these types of geometries have been studied in the literature for the flow conditions expected to be utilized in the aerosol delivery system. Removal of particles is primarily due to impaction and interception on surfaces, as well as gravitational settling. Impaction occurs when the trajectory of a particle drives the particle into a surface, while interception is when a particle deposits onto a surface because its trajectory is less than the particle radius from the surface. This distinction can be seen in Figure 2.3.

**Mixing Chamber**

Aerosol losses in a mixing chamber are a result of particles depositing on the walls of the chamber and particles settling out due to gravitational forces. Particle deposition onto walls can occur due to diffusion, turbulence or body forces such as centripetal force. For vessels in a laminar flow regime, gravitational sedimentation is the major driving force behind the removal of particles.

Corner and Pendlebury [35] first studied aerosol losses in a rectangular box due to turbulent diffusion and gravitational sedimentation. Crump and Seinfeld [36] expanded on this study and developed an equation to determine the aerosol loss in a vessel of any shape for mono-disperse aerosols. Park et al. [37] used the modal method to account for the increased aerosol losses of a polydisperse aerosol compared to a monodisperse aerosol with the same mean diameter.

In a cyclone-type chamber, the rotating flow will also contribute to particle removal. Large particles having more inertia drift into the wall of the chamber and are removed from suspension. This mechanism has been studied by by Fuchs [38] and...
Friedlander [32].

Straight Tubes

Similar to mixing chambers, removal of aerosol particles in a straight tube is due to turbulent diffusion of particles onto the walls and gravitational settling.

Aerosol loss by sedimentation in straight tubes was reported by Thomas [39] and Fuchs [38]. Heyder and Gebhart [40] expanded on this work to develop a model that could be used for straight tubes on an incline. Turbulent deposition in a tube was investigated by Friedlander and Johnstone [41] as well as Davies [42].

The simultaneous modeling of these two effects, sedimentation and turbulent diffusion, was presented by Anand and McFarland [43]. In their study a method of determining the optimal diameter of a tube for a particular particle size and volumetric flow was also developed. When the tube diameter is below the optimal, turbulent diffusion results in large aerosol losses. When the tube diameter is larger than optimal, particles are not kept in suspension as well and particles sediment out due to gravitational forces.

Tube Bends

Particle loss in tube bends has been studied by many groups. These studies have focused on the velocity profile of the flow in the bend and exclude effects of gravity. Early studies by Landahl and Herrmann [44], Hacker et al. [45] and Yeh [46] assumed parabolic, potential and laminar plug flows respectively, through a two dimensional channel bend. Cheng and Wang [47] studied this phenomenon using a two dimensional forced vortex flow in a pipe with circular cross-section. None of these studies included the secondary fluid motion that develops as the gas moves through a bend, and thus the authors’ formulations tend to overpredict experimental results.

Crane and Evans [48] were the first to include the effects of secondary flow using the numerical technique of Rowe [49]. However, their results did not differ markedly from the previous studies. Cheng and Wang [50] took another look at the problem using the developed laminar flow field of a bend proposed by Mori and Nakayama [51]. Their results showed a much better fit to data that had been collected by Johnston et al. [52] for a
Reynolds number of \( \approx 1000 \).

Tsai and Pui [53] further investigated the deposition for developing flow using a three-dimensional, fully elliptic finite difference program developed at the University of Minnesota. They obtained results similar to Cheng and Wang [50] and showed that deposition is not consistent throughout the bend.

Important non-dimensional parameters used in tube bend models include the curvature ratio

\[
R_o = \frac{r_b}{r_{tu}}
\]

(2.16)

the Dean number

\[
Dn = \frac{Re}{\sqrt{R_o}}
\]

(2.17)

and the Stokes number where the characteristic distance is the radius of the tube, \( r_{tu} \). In Eq. (2.16), \( r_b \) is the centerline radius of a curved tube, and in Eq. (2.17), \( Re \) is the Reynolds number

\[
Re = \frac{\bar{U}L_{char}}{\nu_g}
\]

(2.18)

with \( \nu_g \) the kinematic viscosity, \( \bar{U} \) the mean axial velocity and \( L_{char} \) a characteristic length, generally taken as the tube diameter.

Valve

The valve in the valve housing provides two obstructions where aerosol particles may be lost. The first obstruction is the cylinder of the valve stem and the second is the disc at the end of the valve. Both of these geometries have been studied in the literature.

In the aerosol system, the flow is at an angle to the cylinder; however, no models were found in the literature for this configuration. Models have been developed for the deposition of aerosol particles in cross-flow with a cylinder and are presented in multiple texts [1, 32, 38].

The theory was originally developed to understand the collection efficiency of filters, which are made up of numerous fibers having the general shape of cylinders. Wessel and Righi [54] expanded on this early work and developed generalized correlations for the
impaction of aerosol particles on cylinders using finite difference methods which solve the equations of motion. Their results were in good agreement with experimental data.

As discussed earlier, the Stokes number is an important non-dimensional parameter when an obstruction is present in an aerosol flow. For cylinders, a critical Stokes number of 0.125 exists below which particles will not be removed [38]. The characteristic distance used for the Stokes number is the cylinder radius.

Ranz and Wong [55] were the first to propose a theoretical relationship based on potential flow for the deposition efficiency of a disc perpendicular to an aerosol flow. Both Gregory and Stedman [56] and May and Clifford [57] conducted experiments to measure the deposition efficiencies. The model proposed by Ranz and Wong [55] followed the same trend as the experimental data, but overpredicted the deposition efficiency for a particular Stokes number.

Vincent and Humphries [58] modified the equation of Ranz and Wong [55] after studying the flow field visualizations captured by May and Clifford [57]. Their modification produced similar results, with deposition efficiencies only slightly lower than Ranz and Wong [55]. Vincent and Humphries [58] proposed that the discrepancy from experimental results might be due to the effect of gravity, which is not taken into account in the model. The experiment by May and Clifford [57] measured deposition of upward flow onto a disc, where gravity could reduce collection efficiency.

Petkovic [59] extended the model of Ranz and Wong [55] to include the effect of gravity on the deposition of a upward or downward flowing aerosol on a disc. However, deposition efficiency curves were not presented to compare the theory to the experimental data.

A validated model for the deposition of horizontally flowing aerosol particles onto a disc as is used in the fuel delivery system was not found in the literature.

2.4 Methods of Solving the GDE

The general dynamics equation is non-linear and therefore, numerical methods must be used to obtain a solution. Methods historically employed can be classified by how the
droplet distribution is approximated. Ones that have been proposed and developed in the literature include continuous, sectional, modal and piecewise. A description of each approach and its development, as well as the advantages and disadvantages are discussed in the following sub-sections.

2.4.1 Continuous

The continuous method utilizes a continuous size spectrum for the DSD function while solving for mechanisms using finite element methods. Suck and Brock [60] developed a model using a J-space transformation that looked at the effect of Brownian coagulation on atmospheric aerosols. Tsang and Brock [61] investigated effects of condensation and evaporation on a DSD. Their model utilized separate techniques to solve for condensation and evaporation which were later coupled by Tsang and Brock [62] so that both coagulation and evaporation could alter the DSD within a particular simulation. Tsang and Brock [63] also developed a model combining their previous work that solved for coagulation, condensation and evaporation.

Continuous methods can be constructed to produce solutions to any desired level of accuracy and are useful when studying individual mechanisms. However, the computational demands of these accurate solutions are very large [64] and therefore, the continuous method is not used in present day aerosol studies.

2.4.2 Modal

The modal method was first introduced by K. T. Whitby [65] to study aerosol growth rates in the atmosphere. A detailed description of the method and derivation of important equations was presented by E. R. Whitby and McMurry [66].

The modal method treats the aerosol DSD as a combination of distinct aerosol populations each with its own parameterized distribution function. Each function is referred to as a mode and generally up to three modes can be used in simulations. The major assumption of the modal method is that the initial and final DSDs can be represented by the same parameterized distribution function. The most common distribution function is the log normal distribution, but others are used as well [67].
inputs and outputs of a modal model that define the DSD are parameters that describe the modes of the aerosol population.

The modal method is a computationally efficient approach for tracking aerosol dynamics and is most appropriate for large scale applications where accuracy is not critical [64]. The method is fairly accurate for dilute aerosols, but is challenged when aerosol particle densities are large because it overpredicts coagulation rates due to the fixed distribution function [68]. The need for the aerosol DSD to be represented by a distribution function also limits use of the method as it is not able to accurately track droplet populations that deviate from the specified function. Some efforts have been made to correct this problem and improve the accuracy of the modal method, however these have not been adequately validated for a range of conditions [67].

2.4.3 Sectional

The sectional method was first rigorously derived by Gelbard, et al. [69] for a single component aerosol undergoing coagulation. The derivation was expanded by Gelbard and Seinfeld [70] for multi-component aerosols undergoing coagulation, intra-particle chemical reaction, gas to particle conversion as well as particle source and removal mechanisms.

The sectional method approximates the aerosol DSD by dividing it into different size classes, or sections, where aerosol particles are assumed to be the same size. The sections are classified by a property of the aerosol that is conserved during coagulation (e.g., mass). The number of sections used is an important factor in all sectional models; as the number of sections in a model increases, the results become closer to the solution when a continuous distribution is used.

When a sufficient number of ‘bins’ is used in a model, the sectional method conserves particle count and total mass when handling the mechanisms of coagulation and removal. However, this is not the case for growth mechanisms. This is because the particles grow in amounts that can be smaller than the bin spacing. One- and two-degrees of freedom (DOF) methods have been developed to handle the growth mechanism.

According to Makiharju [71], the 1 DOF method is more commonly used in aerosol modeling and is relatively easy to comprehend and implement. With this method, one
parameter is tracked and conserved during a simulation, most commonly the mass of aerosol particles. This method provides accurate results as long as a sufficient number of bins is used to control numerical diffusion. However, this requirement increases the computational demand of the model.

A 2 DOF model follows and conserves two parameters of the aerosol particles [71, 72]. The two parameters are generally the mass and number of the aerosol particles. The method provides results that are comparable to the 1 DOF method with greater efficiency. However, this method is not as straightforward as the one DOF method and requires filtering and other special handling of bins to overcome non-physical results that can be generated by the model.

The sectional method allows a model developer to tailor the trade off between accuracy and speed based on the application. The methodology is well suited to track rapidly changing DSDs [69] as there is no need to conform to a particular DSD profile, but the computational demands are larger than modal methods [68]. When the section sizes are appropriate, the sectional method is an accurate means of tracking the dynamics of an aerosol DSD. It is the most common method used in aerosol models [73].

2.4.4 Piecewise

The piecewise method is a hybrid of the modal and sectional methods. In this approach, the aerosol DSD is divided into sections, and the distribution in each of the sections is represented by an analytical function. Log normal distributions are utilized by von Salzen [73] to represent the aerosol population in each section while Sandu [74] uses polynomial representations of varying degree.

The piecewise method is relatively new and has not seen much utilization or validation in aerosol dynamics models. Both studies [73, 74] demonstrate that accurate results can be obtained more efficiently when compared to sectional and modal methods. However, both sets of researchers also acknowledge that more thorough investigations looking into the generally applicability of the method needs to be completed before the piecewise method can be widely adopted.
Based on the motivation and objectives of the current study, a sectional method was selected as the framework for solving the GDE and the evolution of the fuel aerosol within the aerosol fueling system.
Chapter 3

AEROSOL MODEL IMPLEMENTATION

A model was developed to determine the aerosol delivery efficiency of a system designed to prepare and introduce fuel aerosols into a reaction chamber of an RCM. As mentioned earlier, the aerosol delivery system is composed of a mixing chamber, valve housing and a reaction chamber with a delivery tube connecting the mixing chamber and valve housing, a schematic of which is shown in Figure 1.12. Aerosols are desired which have initial mixture equivalence ratios from 0.25 to 2 at 5 - 21% O_2 and are delivered with reaction chamber Reynolds numbers between 500 and 800, which has been found to prevent bulk scale aerosol stratification [28].

At these delivery conditions the mechanisms directly and indirectly accounting for aerosol loss are evaporation, coagulation, deposition and sedimentation. Evaporation and coagulation models apply to all sections of the delivery system. Deposition and sedimentation mechanisms are contained within removal sub-models. All of the models employed will be discussed later in the chapter.

One goal of the aerosol model is to be able to determine how various fuel and gas combinations behave in a delivery system. Specifically, what fuel concentrations can be reliably delivered and the locations where the fuel is removed from suspension. To do this utilizing high fidelity CFD modeling can be very time consuming and this does not allow for a variety of fuels, gases, operating conditions or system configurations to be tested easily. Therefore a model using the bulk properties of the aerosol was developed.

Based on the review of methods used to predict aerosol behavior in Chapter 2, the 1 DOF sectional method was selected for the basis of the model. This method provides fairly accurate results and is proven to be dependable. One downside is that the DSD
Figure 3.1: Schematic of aerosol fueling system as depicted in the model. Inputs, outputs and example control volumes of the aerosol model are indicated. The plot shows an example sectional droplet distribution.

needs to be divided into many sections in order to limit diffusion. A discussion of the bin sizing used in the aerosol model can be found in Appendix B.

Similar to the DSD, the aerosol system needs to be divided into multiple control volumes in order to capture the effects of the interacting mechanisms. A diagram showing the inputs into the model, where each sub-model is used, and the outputs of the model can be seen in Figure 3.1. A discussion of how the system components are discretized can also be found in Appendix B.

The model assumes 1D, laminar flow and also assumes bulk homogeneity within each discretized control volume. This means that the droplet distribution, flow rate and properties, as well as gas and liquid temperatures are uniform throughout a cross section, of the reaction chamber, for example. Energy addition to the system is taken into account by employing a constant wall temperature and a model for bulk heat transfer.

In describing the sub-models, liquid refers to fuel droplets, vapor refers to fuel in the vapor phase, and gas refers to the bath gas initially introduced (e.g, air).

3.1 Main Model

Inputs into the model include a droplet distribution, evaporation coefficient (discussed in Appendix B), properties of the bath gas and fuel, ambient conditions, geometries of the
system components, the size in kmol of the smallest particle, equivalence ratio of the aerosol created in the mixing chamber ($\phi_{in}$) and the Reynolds number of the reaction chamber ($\text{Re}_{RC}$).

The thermophysical properties of the gas and fuel are determined initially and then updated after every time step. The thermophysical models used for these properties are reviewed in Appendix A. Most models are chosen based on the recommendations from Poling, et al. [75].

The mass flow rate of the liquid from the aerosol generator and the bath gas are specified from the desired $\text{Re}_{RC}$ and $\phi_{in}$ as well as the associated temperature pressure, etc. and are used throughout the model to determine the progression of the aerosol through the system. Discretized volumes of the system components are calculated and used to determine retention times. The initial mass of fuel per kg of bath gas is calculated and is used to determine the percent loss in each of the system components.

The model handles each component of the delivery system in a similar manner. The evaporation sub-model generally limits the time step, and the sub-models are implemented in the following order: evaporation, energy balance, coagulation and removal. When the coagulation sub-model limits the time step, the implementation is: coagulation, energy balance and removal. The sub-models and time step control are discussed in the remainder of this chapter.

The mass of the aerosol liquid phase is calculated as

$$m_l = \sum_{j=1}^{X} n_j N_j M_p,$$  \hspace{1cm} (3.1)

where $X$ is the total number of DSD sectional bins, $n_j$ is the molar particle size of bin $j$, $N_j$ is the particle concentration in bin $j$ and $M_p$ is the molar mass of the particle. The mass of the vapor phase is initially calculated using the ideal gas law

$$m_v = \frac{P_\infty V M_p}{R_u T}$$  \hspace{1cm} (3.2)

where $V$ is the discretized volume size and $R_u$ is the universal gas constant. Thereafter it
Figure 3.2: Energy fluxes in a control volume.

is calculated in the evaporation sub-model, discussed in Section 3.3.

The loss in a component of the fuel system is determined by

\[ L = \frac{(m_l + m_v)_\text{in} - (m_l + m_v)_e}{(m_l + m_v)_0}. \] (3.3)

where subscripts \( \text{in} \), \( e \), and 0 represent the inlet, exit and \( t = 0 \) values. The overall loss in the system is defined as

\[ L_{\text{sys}} = L_{mc} + L_{dt} + L_{vh} + L_{rc}. \] (3.4)

3.2 Energy Balance

During each time step, the energy flows are taken into account using the bulk properties of the aerosol components (i.e., the liquid and vapor phases of the “fuel” as well as the diluent gas). The mass of the vapor phase is tracked in the aerosol model and the mass of the liquid is calculated using the DSD. The gas phase (i.e. oxidizer and diluent) does not change mass during a simulation and the pressure increase due to vaporization is not included due to the small contribution to total pressure.

A schematic of the energy flows in a discretized control volume can be seen in Figure 3.2. While the aerosol passes through the control volume, energy is added via convection from the walls and transferred from the gas to liquid phase. Energy also moves to the vapor phase as the liquid evaporates. The temperature of the aerosol in the control volume is assumed to be homogeneous and the exit temperature is determined by solving
the following energy balance

\[
[\dot{m}_l h_l(T) + \dot{m}_v h_v(T) + \dot{m}_g h_g(T)]_e
\]

\[
= \dot{q}_w + [\dot{m}_l h_l(T) + \dot{m}_v h_v(T) + \dot{m}_g h_g(T)]_in
\]

where \(\dot{q}_w\) is the energy flux from the wall into the control volume, \(\dot{m}\) is the mass flux and \(h\) is the enthalpy. It is assumed here that the rate of heat transfer from the gas to liquid phase to supply the required vaporization enthalpy is infinitely fast so that the gas and liquid phases can be assumed to be in thermal equilibrium. The approximation used is appropriate because the Lewis numbers of the liquids of interest are relatively high and the relative time scale on which evaporation occurs is fairly small. The Lewis number represents the ratio of thermal diffusivity to the mass diffusivity and the values are approximately 1 for water and 4 for large hydrocarbons (i.e. 12 - 18 carbon atoms). Thus, heat and mass transfer occur on roughly the same time scale for water which, for the initial evaporating in the mixing chamber, is on the order of \(10^{-5}\) s; for hydrocarbons of interest heat transfer is \(\approx 4\) times faster than the diffusion flow of fuel vapor from the droplets. These time scales are very small in comparison to the retention time of the aerosol in the fuel delivery system which is on the order of 10 s. Therefore, during nearly all of the time in the system, the gas and liquid phases are expected to be nearly the same temperature.

This homogeneous temperature approximation could be improved by incorporating a diffusion limited rate equation for the heat transport while tracking the liquid and gas phase temperatures through the delivery system. Accounting for liquid phase thermal diffusion though would also increase the complexity of the model.

The energy flux from the wall during a time step is determined by assuming a constant wall temperature and employing Newton’s law of cooling

\[
\dot{q}_w = h_{conv} A_w (T_w - T)
\]

where \(A_w\) is the surface area of the wall in the control volume. The convection coefficient
Table 3.1: Coefficients for use with Eq. (3.9)

<table>
<thead>
<tr>
<th>$z$</th>
<th>0</th>
<th>1</th>
<th>2</th>
<th>3</th>
<th>4</th>
</tr>
</thead>
<tbody>
<tr>
<td>$G_z$</td>
<td>0.749</td>
<td>0.544</td>
<td>0.463</td>
<td>0.415</td>
<td>0.383</td>
</tr>
<tr>
<td>$\lambda_z$</td>
<td>7.313</td>
<td>44.61</td>
<td>113.9</td>
<td>215.2</td>
<td>348.6</td>
</tr>
</tbody>
</table>

is calculated by

$$h_{conv} = \frac{Nu k_g}{L_{char}}$$

(3.7)

where $k_g$ is the thermal conductivity of the gas and $Nu$ is the local or average Nusselt number.

Inside the mixing chamber, the Nusselt number is determined using the model for a cyclone proposed by Szekely and Carr [76]

$$Nu = 0.042Re^{0.8}$$

(3.8)

In straight tubes, the Nusselt number is calculated from the internal developing flow model using the first five components of the infinite series [77]

$$Nu = \frac{\sum_{z=0}^{\infty} G_z e^{-\lambda_z x^+}}{2\sum_{z=0}^{\infty} G_z/\lambda_z e^{-\lambda_z x^+}}$$

(3.9)

where $G_z$ and $\lambda_z$ are the eigenvalues of the solution where these are listed in Table 3.1, and

$$x^+ = \frac{2x^*}{RePr}$$

(3.10)

with $x^*$ being the axial distance along a length of tube non-dimensionalized by the total length of the tube (i.e. $x/x_{tu}$).

In tube bends, the Nusselt number is determined using the a fully developed flow model given by Kalb and Seader [78]

$$Nu = 0.836Dn^{0.5}Pr^{0.1}$$

for $Dn \geq 80, \ 0.7 < Pr < 5$

(3.11)

where $Dn$ is the Dean number as calculated in Eq. (2.17).
In the previous equations the Reynolds number is calculated using Eq. (2.18) and the Prandtl number is calculated as

$$Pr = \frac{C_p \mu g}{k_g} \quad (3.12)$$

where $C_p$ is the specific heat capacity. The average velocity in the mixing chamber is calculated from

$$\bar{U} = \frac{\dot{m}_g}{\rho_g x^2} \quad (3.13)$$

where $x$ is the cyclone radius or the smallest dimension of the box. In all other components, the average velocity in each section is

$$\bar{U} = \frac{\dot{m}_g}{\rho_g A_{cs}} \quad (3.14)$$

where $A_{cs}$ is the cross sectional area of a component section.

### 3.3 Evaporation

In using the sectional method to track the aerosol dynamics, the biggest challenge is the incorporation of an evaporation model. This is because droplet sizes decrease on a continuum, and discretizing the continuum (i.e. droplet volume) leads to an acceleration of the process. The algorithm utilized for the evaporation sub-model is the 1 DOF method described by Moffat [72] where

$$\frac{dQ_j}{dt} = R_j \left(1 + \frac{n_{j-1}}{n_j - n_{j-1}}\right) - \frac{R_{j+1}n_j}{n_{j+1} - n_j} \quad (3.15)$$

where $Q_j$ is the total concentration of liquid molecules in bin $j$. The net flux of the concentration of liquid molecules into a bin is

$$R_j = \frac{q_j}{n_j} \int_{n_j}^{n_{j+1}} E(n) dn \quad (3.16)$$
where \( E(n) \) is the molar evaporation rate and the molecule concentration of particles with size \( n_j \) is

\[
q_j = \frac{n_jN_j}{n_j - n_{j-1}} \quad (3.17)
\]

Any particle growth model, or in this case shrinkage, may be incorporated into the algorithm. The one used for the aerosol model is given by Davies [33]

\[
E(n) = \frac{2\pi d_p D_v M_p f(\alpha)f(\text{Re}, \text{Sc}) (P_\infty - P_{sat})}{R_u T} \quad (3.18)
\]

where

\[
d_p = \left( \frac{n M_p b}{\pi \rho_p} \right)^{1/3} \quad (3.19)
\]

A positive value of \( E \) represents particle growth (i.e. condensation) and a negative value is particle shrinkage (i.e. evaporation). As stated earlier, a spatially homogeneous vapor pressure is assumed for the far field, while the vapor just above the surface of the droplet surface is assumed to be at the saturated condition. The gradient between the two concentrations drives the evaporation process.

In Eq. (3.18), \( f(\alpha) \) is the correction as the particle size approaches the mean free path of the vapor, \( \lambda_v \). The correction used in the aerosol model was proposed by Fuchs and Sutugin [34]

\[
f(\alpha) = \frac{0.75\alpha(1 + \text{Kn})}{\text{Kn}^2 + \text{Kn} + 0.283\text{Kn}\alpha + 0.75\alpha} \quad (3.20)
\]

with the accommodation coefficient, \( \alpha \), set to 1 in the aerosol model. The Knudsen number is defined in Eq. (2.5), however the mean free path of the vapor is defined differently as

\[
\lambda_v = \frac{3D_v}{\bar{c}} \quad (3.21)
\]

where the mean speed of the vapor molecules, \( \bar{c} \), is

\[
\bar{c} = \left( \frac{8k_b T A_v}{\pi M_v} \right)^{1/2} \quad (3.22)
\]

with \( A_v \) representing Avogadro’s number.
The other correction function in Eq. (3.18) is due to relative motion between particles and is presented by Davies [33]

\[ f(Re_p, Sc) = 1 + 0.276Re_p^{1/2}Sc^{1/3} \]  

(3.23)

where the Schmidt number, \( Sc \), is \( \nu_g/D_v \).

The new DSD and total mass of the vapor phase are calculated as

\[ N_j = q_j(n_j - n_{j-1}) + \frac{dQ_j}{dt} \Delta t \]  

(3.24)

and

\[ m_{v,e} = m_{v,in} + \sum_{j=1}^{X} n_j M_p (N_{j,in} - N_{j,e}). \]  

(3.25)

The new vapor pressure of the far field is determined using the ideal gas equation of state, Eq (3.2).

### 3.4 Coagulation

The coagulation algorithm in the aerosol model is modified from Mäkiharju [71] and is presented by Pirjola et al. [79] where the rate of droplet change is given by

\[
\frac{dN_i}{dt} = \sum_{i=1}^{i} \sum_{k=1}^{i} K_{j,k} \frac{N_j(t)N_k(t)}{1 + \delta_{j,k}} \left( \frac{N_j(t)N_k(t)}{n_i - n_{i-1}} \delta_{n_j+n_k,|n_i-1,n_i|} ight) \\
+ \sum_{i=1}^{i} \sum_{k=1}^{i} K_{j,k} \frac{N_j(t)N_k(t)}{1 + \delta_{j,k}} \left( \frac{n_{i-1} - n_i}{n_{i+1} - n_i} \delta_{n_j+n_k,|n_i-1,n_i|} \right) \\
- N_i(t) \sum_{j=1}^{X} K_{i,j} N_j(t)
\]

(3.26)

with \( K_{j,k} \) the combined coagulation coefficient between particles in bin \( j \) and \( k \) and Kronecker’s deltas are

\[
\delta_{i,k} = \begin{cases} 
0, & \text{if } j \neq k \\
1, & \text{if } j = k 
\end{cases}
\]  

(3.27)
Table 3.2: Coefficients for use with Eq. (3.32)

<table>
<thead>
<tr>
<th></th>
<th>1</th>
<th>2</th>
<th>3</th>
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<th>5</th>
</tr>
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<tr>
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<tr>
<td>$G_k$</td>
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<td>-</td>
<td>-</td>
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<td>-</td>
</tr>
<tr>
<td>$H_k$</td>
<td>2.843</td>
<td>-</td>
<td>-</td>
<td>-</td>
<td>-</td>
</tr>
</tbody>
</table>

and

$$
\delta_{n_j+n_k|n_i,n_{i+1}} = \begin{cases} 
0, & \text{if } n_j + n_k \notin [n_i, n_{i+1}] \\
1, & \text{if } n_j + n_k \in [n_i, n_{i+1}] 
\end{cases}
$$

Any coagulation kernel may be used in this algorithm. The kernel selected for the aerosol model is presented by Sajo [80, 81] with the Cunningham coefficient added according to Hinds [1] and the collision efficiency, $\epsilon$, included according to Williams [82],

$$K_{i,j} = \gamma_{i,j}\rho g \left[ \frac{2kbT}{3\mu_g} \left( \frac{C_i}{r_i} + \frac{C_j}{r_j} \right) (r_i + r_j) + \frac{2\epsilon g p \pi}{9\mu_g} (r_i + r_j)^2 |C_i r_i^2 - C_j r_j^2| \right]$$

with the first and second terms in the bracket representing Brownian and gravitational coagulation, respectively and

$$\epsilon = 1 + \frac{1 + p^2}{2(1 + p)^3} - \frac{3}{2(1 + p)}$$

where

$$p = \frac{\min(r_i, r_j)}{\max(r_i, r_j)}$$

The correction factor for the sum coagulation kernel for Brownian and gravitational
coagulation is
\[
\gamma_{i,j} = \begin{cases} 
\sum_k A_k e^{-k\beta_{i,j}} + \sum_k B_k \beta_{i,j}^k + C_1, & \text{if } \beta_{i,j} \leq 5.5 \\
\sum_k D_k [\ln(\beta_{i,j})]^k + \sum_k E_k \beta_{i,j}^{k/3} + \sum_k F_k \beta_{i,j}^{k/3} + G_1, & \text{if } 5.5 < \beta_{i,j} \leq 2000 \\
1 + H_1 \beta_{i,j}^{-2/3}, & \text{if } \beta_{i,j} > 2000
\end{cases}
\] (3.32)

where the coefficients are listed in Table 3.2 and \(\beta_{i,j}\) is a dimensionless quantity indicating the relative importance of diffusion versus gravitational settling and is calculated by
\[
\beta_{i,j} = \frac{4\pi \rho_p g (r_i + r_j) \left| C_i r_i^2 - C_j r_j^2 \right|}{3 k_b T \left( \frac{C_i}{r_i} + \frac{C_j}{r_j} \right)}
\] (3.33)

The new DSD is calculated by
\[
N_{i,e} = N_{i,in} + \frac{dN_i}{dt} \Delta t
\] (3.34)

### 3.5 Removal Models

Sub-models for removal are described in this section. Sub-models for the valve stem and seat are implemented after the last step of the valve housing, while other removal models are used in each control volume within the system. For each sub-model the exiting droplet concentration in a bin is
\[
N_{i,e} = (1 - \chi_i) N_{i,in}
\] (3.35)

where \(\chi\) is the particle fraction loss in a component, or a particular section of a component. Since the removal of particles is dependent on droplet size, each bin will have a different value for \(\chi\).

**Mixing Chamber Removal Model**

The removal modeling of the mixing chamber is handled differently for the box and cyclone mixing chambers because the configurations and thus internal flow dynamics are different. The box-type mixing chamber introduces the bath gas at the bottom and the
Figure 3.3: (a) Flow field for the cyclone [83] and (b) particle traces for the box [28] designs of the mixing chamber. The flow in the box is non-uniform while the flow in the cylinder is controlled.

liquid droplets at the top with the aerosol exit at the top. The cyclone mixing chamber, on the other hand, introduces both the bath gas and the droplets at the top of the vessel while aerosol exits at the bottom. Simulations of the fluid dynamics within the box and cyclone, seen in Figure 3.3, illustrate the non-uniform behavior in the box configuration. The sub-model for the cyclone will be discussed first, followed by the sub-model for the box mixing chamber.

The cyclone mixing chamber deposition sub-model uses two models from the literature. Droplet loss due to gravitational settling and diffusion is calculated using the model developed by Crump and Seinfeld [36]. The removal rate in a stirred tank is defined as

\[
\chi_{mc} = \frac{y \sin(\pi / y)(k_e D_p^{y-1})^{1/y} A_{mc, w}}{\pi A_{mc, bo} h_{mc}} + \frac{U T_S}{h_{mc}} \coth \left( \frac{\pi U ts}{2y \sin(\pi / y)(k_e D_p^{y-1})^{1/y}} \right) \quad (3.36)
\]

where \( A_{bo} \) is the area of the bottom of the mixing chamber, \( y \) is a constant constrained to a value between 2 and 3, with the value set to 2.7 in the present simulations, and the
coefficient of the eddy diffusivity defined as

\[ k_e = 2.87 \times 10^2 (\bar{U})^{9/5} k_o^2; \]  

(3.37)

with \( \bar{U} \) in m/s and the von Karman constant, \( k_o \), equal to 0.4.

Droplet loss due to the rotating flow in the cyclone is calculated using the theory presented by Friedlander [32]. The critical distance from the wall where fuel particles will deposit is determined by

\[ z_c = \frac{d_p \rho_p N_t \bar{U}}{\mu g r_{mc}} \]  

(3.38)

where \( N_t \), the number of rotations, is determined by

\[ N_t = \frac{h_{mc} V_{cv}}{r_{mc} V_{mc}} \]  

(3.39)

with \( h_{mc} \) as the height of the mixing chamber and \( V_{cv} \) and \( V_{mc} \) the volumes of the control volume and mixing chamber, respectively. The average velocity is calculated based on

\[ \bar{U} = \frac{\dot{m}_g}{\rho g r_{mc}^2} \]  

(3.40)

Assuming a spatially uniform distribution, the fraction of particles lost is then \( z_c/r_{mc} \).

For the box mixing chamber, the introduction of the gas and droplets at two locations and the complex flow field inside the box make it impossible to accurately simulate aerosol behavior using the bulk model developed here. A more sophisticated CFD formulation is required. For this work, the model of Crump and Seinfeld is utilized with a retention time specified as 70\% of the characteristic retention time. This seems appropriate based on the assumption that the bath gas sweeps half of the volume before mixing with the droplets and once mixed, it might also recirculate. This type of fluid motion is observed in Figure 3.3b and the results predicted by this model fall close to the experimentally measured values.
Straight Tube Removal Model

The straight tube removal sub-model used in the aerosol model was developed by Anand et al. [8, 43]. This sub-model is used for straight tubes within the fueling system as well as the reaction chamber. The particle fraction loss in the tube is

\[ \chi_{tu} = 100 \left( 1 - e^{-\frac{\pi d_{tu} U_{ef} x_{tu}}{Q}} \right) \tag{3.41} \]

where \( Q \) is the volumetric flow, \( x_{tu} \) is the length of the tube segment and the effective velocity is

\[ U_{ef} = \frac{U_{df} \theta_c}{\pi} + \frac{U_{df}}{2} + \frac{U_{TS} \sin(\phi_{tu}) \cos(\theta_c)}{\pi} \tag{3.42} \]

with \( \phi_{tu} \) as the angle of the tube from vertical and the settling velocity, which is dependent on droplet size, calculated from Eq. (2.2). The critical angle in a tube cross section defined as

\[ \theta_c = \begin{cases} 0, & \text{if } U_{TS} \sin(\phi_{tu}) = 0 \\ \arcsin \left( \frac{U_{df}}{U_{TS} \sin(\phi_{tu})} \right), & \text{if } U_{TS} \sin(\phi_{tu}) > U_{df} \\ \pi/2, & \text{if } U_{TS} \sin(\phi_{tu}) \leq U_{df} \end{cases} \tag{3.43} \]

The velocity caused by diffusion is defined as

\[ U_{df} = U_+ \tilde{U} \sqrt{f/2} \tag{3.44} \]

when

\[ U_+ = \begin{cases} 6.9 \times 10^{-4} \tau_+^2, & \text{if } \tau_+ \leq 15 \\ 0.16/\tau_+^{0.086}, & \text{if } \tau_+ > 15 \end{cases} \tag{3.45} \]

and

\[ \tau_+ = \frac{\tau U f Re}{d_{tu}} \tag{3.46} \]
The friction factor is given by the Blasius equation

\[ f = \frac{0.316}{4Re^{0.25}} \]  

(3.47)

where the Reynolds number is based on the tube diameter.

Bend Removal Model

The deposition sub-model developed by Tsai and Pui [53] was initially incorporated into the model in order to determine the removal of aerosol particles, assuming laminar flow. However, this sub-model significantly underpredicts experimentally measured aerosol loss within the system mock up, as well as a simple tube bend. In an attempt to reconcile these discrepancies an empirical correlation was developed based on the Tsai and Pui model, and validated by experimental gravimetric measurements. The bend removal model can be written as

\[ \chi_b = \frac{\theta_b}{90} \chi_{b,LF} + 3.75 \chi_{tu} \]  

(3.48)

Here \( \chi_{b,LF} \) represents a correlation to the Tsai and Pui model which is dependent on droplet Stokes number, and \( \chi_{tu} \) is a gravitational settling component. A detailed discussion of this is included in Appendix C where the gravitational validation measurements are reviewed. The sub-model assumes uniform deposition throughout the bend and was developed over a range of Dean numbers with \( Re_o = 7.38 \). The bend is assumed to be positioned such that the inlet is above a horizontal outlet with the bend angle, \( \theta_b \), measured in degrees.

This combined sub-model is not expected to be completely accurate when used in the aerosol model to predict losses. For example, in the aerosol system the bend angles will be less than 90 degrees and some bends are positioned such that the inlet is below a horizontal outlet. However, the loss in the valve housing in most configurations only contributes a small amount to the total loss (i.e. 4 - 12 %) so inaccuracies of this sub-model do not have a great affect on the loss predictions. Though this is not ideal, it allows for a determination of aerosol behavior in the delivery system over a range of
conditions.

Valve Seat Removal Model

The model for the loss of particles onto the valve seat is taken from Ranz & Wong [55]. Loss onto the surface, which is modeled as a disc obstructing the flow, is determined by

\[ \chi_{di} = \frac{A_{di}}{A_{cs}} \left[ \frac{S_2 - S_1}{S_2 e^{S_2 T_c} - S_1 e^{S_1 T_c}} \right]^2 \]

(3.49)

with \( A_{di} \) as the projected area of the disc, \( A_{cs} \) as the cross-section of the aerosol flow (i.e. the tube cross section) and

\[ S_{1,2} = -\frac{1}{4St} \pm \sqrt{\left( \frac{1}{4St} \right)^2 + \frac{1}{2St}} \]

(3.50)

and the dimensionless time of surface impact is

\[ t^*_c = \frac{\text{atan} \left( \frac{4qSt}{8St-1} \right)}{q} \]

(3.51)

with

\[ q = \sqrt{\frac{1}{St} - \left( \frac{1}{4St} \right)^2} \]

(3.52)

The characteristic length for the determination of the Stokes number in the previous equations is the diameter of the disc.

Valve Stem Removal Model

The model proposed by Wessel and Righi [54] is used to calculate the loss of aerosol particles due to the stem of the valve. The deposition efficiency onto this cylindrical
obstruction is

\[
\frac{\chi_{\text{stem}}}{A_{\text{stem}}/A_{\text{cs}}} = \begin{cases} 
0, & \text{if } \text{St}_e < 0.125 \\
\text{A} \ln(8 \text{St}_e) + \text{B} (\text{St}_e - 0.125) + \text{C} (\text{St}_e - 0.125)^2, & \text{if } 0.125 < \text{St}_e \leq 0.5 \\
[1 + \text{D} (\text{St}_e - 0.125)^{-1} + \text{E} (\text{St}_e - 0.125)^{-2} + \text{F} (\text{St}_e - 0.125)^{-3}]^{-1}, & \text{if } 0.5 < \text{St}_e
\end{cases}
\]

(3.53)

where the coefficients are listed in Table 3.3 and

\[
\text{St}_e = \text{St} \psi
\]

(3.54)

with

\[
\psi = \frac{3 \left[ \sqrt{c \text{Re}_p^{1/3}} - \text{atan}(\sqrt{c \text{Re}_p^{1/3}}) \right]}{c^{3/2} \text{Re}_p}
\]

(3.55)

where \( c = 0.158 \) and \( \text{Re}_p \) is the Reynolds number with the particle diameter as the characteristic length and \( \bar{U} \) as the average gas velocity.

### 3.6 Time Step Control

Within the aerosol model the time step must be controlled in order to prevent numerical diffusion as droplets are moved between bins due to evaporation and coagulation, and control the accuracy of the application of the removal sub-models. The size of time steps in the aerosol model, and therefore the bulk transport within the system components, is modulated by the evaporation and coagulation sub-models. This is conducted in order to prevent bins from having a negative value for the particle count and to improve the resolution of the model.

The evaporation sub-model is used to set the time step when the pressure of the

<table>
<thead>
<tr>
<th>A</th>
<th>B</th>
<th>C</th>
<th>D</th>
<th>E</th>
<th>F</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.01978749</td>
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<td>-0.0482858</td>
<td>1.54424</td>
<td>-0.538013</td>
<td>0.2020116</td>
</tr>
</tbody>
</table>
vapor phase is less than the saturation pressure. The number of molecules allowed to evaporate is determined by

\[ n_{\text{evap}} = \frac{V(P_{\text{sat}} - P_{\infty})}{R_u T} \]  (3.56)

when

\[ P_{\infty} \geq (1 - 2c_{\text{evap}})P_{\text{sat}} \]  (3.57)

where the value \( c_{\text{evap}} \) is used to control the size of the time steps during evaporation and has a value set to 0.02 in the aerosol model. For an explanation of how this number was chosen refer to Appendix B.

When the condition in Eq. 3.57 is not met, the number of molecules allowed to evaporate is determined by

\[ n_{\text{evap}} = c_{\text{evap}} \frac{P_{\text{sat}} V}{R_u T} \]  (3.58)

The time step is first set by

\[ t_{\text{step}} = -\frac{n_{\text{vap}}}{\sum_{j=1}^{X} R_j} \]  (3.59)

where the net particle concentration flux, \( R_j \), is calculated using Eq. (3.16). If necessary, the time step is decreased by half until the minimum value of the particle concentration in all bins is \( \geq 0 \).

When the vapor phase is saturated (i.e. \( P_{\infty} = P_{\text{sat}} \)), the evaporation sub-model is not utilized and the coagulation sub-model is used to determine the time step.

The maximum time step within a component is determined by dividing the component into discrete volumes and then calculating the retention time of one section based on \( \bar{U} \). If necessary, the time step is reduced in the same manner as the evaporation sub-model.

The number of discretizations used for the mixing chamber is 60. All other components use a value of 16. Refer to Appendix B for the rationale behind these numbers. Using the maximum time step as an initial value, the time step is then determined the same way as in the evaporation sub-model.
In both sub-models, the cumulative volume swept by the aerosol is determined by

\[ V_{tot,e} = V_{tot,in} + \frac{\dot{m}_g \Delta t}{\rho_g}. \]  

(3.60)

where \( V_{tot,in} \) is the volume swept before entering the control volume. If the cumulative volume swept is greater than the total volume of the component, the time step is reset so that the cumulative volume swept is equal to the total volume of the component.

The next chapter presents validation results for the model based on experimental data acquired from a mock-up of an aerosol RCM.
RESULTS AND DISCUSSION

In this chapter, the aerosol model described in the Chapter 3 is first validated using an aerosol RCM mock up with water as the ‘fuel’ and the model is then utilized to gain insight into the behavior of the system. Of particular interest is how the aerosol evolves as it is transported through the system components, which mechanisms are most important during typical operation and how the operating conditions, including the fuel and diluent conditions, impact the system performance. System performance is assessed here based on aerosol loss and peak droplet diameter in the reaction chamber. The chapter concludes with a discussion of potential improvements to the delivery system and aerosol model.

4.1 Aerosol System Experiments

Experiments have been conducted to investigate the behavior of an aerosol in an RCM mock up within the Energy Lab at Marquette University. The intent is to understand how various configurations and operating parameters affect system performance. The goal is to

Figure 4.1: Diagram of the aerosol RCM mock up at Marquette University
design a system that can achieve low levels of droplet fallout and bulk scale droplet stratification at operating conditions where stoichiometric fuel loading is possible at atmospheric oxygen concentrations. The RCM mock up was designed, built, and modified by John Bustamante, Mike Denk, Mike Johnson, Alex Polley and Colin Banyon under the direction of Dr. S. Scott Goldsborough. As discussed in previous chapters, the delivery system consists of a mixing chamber, delivery tube, valve housing, reaction chamber and exit manifold. A schematic was presented earlier in Figure 1.12 and is shown again in Figure 4.1. A description of the initial configuration and experiment results follows. As mentioned in Chapter 1, the poor performance seen in the results was a motivation for the current work.

The droplets for the aerosol are generated with tap water using an array of Aeroneb OnQ nebulizers (usually four) that are assembled onto a detachable tray with a covered trough that seats onto the top of the mixing chamber. The initial rectangular mixing chamber measures $0.178 \times 0.152 \times 0.203$ m and is fabricated from 6.4 mm thick acrylic plastic. Air from the building’s compressed air line is introduced at the side of the mixing chamber. The gas flow is metered using orifices from O’Keefe Controls Company. The air pressure before and after the orifice is measured using MGA-100-A-9V digital gages from SSI Technologies, Inc.

After the mixing chamber, the aerosol is transported through a delivery tube that connects to the valve housing. The tubing is Kuri Tec Clearbraid which has an internal diameter (ID) of 12.7 mm and length of 1.65 m.

The initial valve housing (#1) is machined from aluminum and contains a 12.7 mm ID tube which leads to a horizontal cylinder (i.e., the reaction chamber) that is 0.051 m in diameter by 0.038 m long. Inside the valve housing, the path of the tube comes to a right angle after which the aerosol is introduced to the cylinder. At the open end of the valve housing cylinder a 0.064 m diameter by 0.013 m deep counter bore is employed to accept the reaction chamber. This is illustrated in Figure 4.2a.

The reaction chamber is simulated by a cylindrical glass tube with inside diameter of 0.051 m and length of 0.305 m. After the reaction chamber, the aerosol flows through an exit manifold that has the same configuration as the inlet valve housing (#1) and
allows the aerosol to exit the delivery system.

Experiments are conducted at $T \approx 297$ K and $P = 1$ bar. To begin an experiment, the mixing chamber, delivery tube, valve housing, reaction chamber and exit manifold are weighed using a Mettler Toledo SB16001 scale and then assembled. About 200 mL of water is added to a 600 mL beaker and covered to prevent evaporation loss. The beaker, nebulizer tray and both covers are weighed together before the experiment.

The pressure behind the orifice is measured and the mass flow rate of the gas is calculated using the ideal gas, choked flow expression,

$$
\dot{m} = A_o C_d P_{tu} \sqrt{\frac{\gamma}{RT_{tu}}} \left( \frac{2}{\gamma + 1} \right)^{\frac{\gamma+1}{2(\gamma-1)}}
$$

(4.1)

where $A_o$ is the area of the orifice opening, $P_{tu}$ the pressure of the gas behind the orifice, $T_{tu}$ the temperature of the gas behind the orifice, $\gamma$ the specific heat ratio and 0.9 is used as the discharge coefficient, $C_d$ [84].

The water is poured from the beaker into the nebulizer tray and both components are covered to prevent evaporation loss. The nebulizers are powered on to initiate the experiment. The nebulizers run for $\approx 30$ minutes and are then powered off. Adequate flow is required in order to achieve resolution with the Mettler scale. The air flow is allowed to continue until the aerosol clears the system.
Table 4.1: Operating conditions for initial aerosol system experiments

<table>
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<tr>
<th>Case</th>
<th>Mixing Chamber Design</th>
<th>Delivery Tube ID (mm)</th>
<th>Delivery Tube Length (m), Position</th>
<th>Valve Housing Design</th>
<th>Barb</th>
<th>Valve</th>
<th>Water Delivered by Nebulizers (g/min)</th>
<th>Flow Rate (L/min)</th>
<th>Temperature (K)</th>
<th>Pressure (bar)</th>
<th>Aerosol Concentration (mL/L)</th>
<th>Total Aerosol Loss (%)</th>
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<td>no</td>
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<td>18.8</td>
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<td>1</td>
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<td>#1</td>
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<td>no</td>
<td>1.31</td>
<td>18.9</td>
<td>298</td>
<td>1</td>
<td>0.069</td>
<td>20.56±1.92</td>
</tr>
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<td>box</td>
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<td>1.65, Horizontal</td>
<td>#1</td>
<td>yes</td>
<td>yes</td>
<td>1.31</td>
<td>18.7</td>
<td>298</td>
<td>1</td>
<td>0.070</td>
<td>24.37±1.92</td>
</tr>
</tbody>
</table>

The air flow is shut off, the system dismantled and each component weighed again. The difference between the final and initial mass is divided by the duration of the run to determine the rate of aerosol loss in the components. Likewise, the difference between the initial and final mass of the nebulizer tray and beaker is divided by the experiment duration to determine the aerosol delivery rate.

Results using the initial system configuration with operating parameters listed in Table 4.1 are presented in Figure 4.2b. The indicated uncertainty is discussed in Appendix D. These tests are conducted to investigate the influence of restrictions within the system. Case 1 uses no poppet valve and no barb to connect the delivery tube. Case 2 has the barb but no valve, and Case 3 has the connecting barb and the poppet valve. As can be seen in Figure 4.2b, when more restrictions are placed in the path of the aerosol, the losses in the system are increased. This is due to two factors. First, the restrictions may changes in the flow of the aerosol which results in increased deposition of particles that do not adjust to the flow before impacting a wall. Second, restrictions can also provide more opportunity for impaction since more surface area may be presented to the aerosol.

Modifications to these components and system configuration were undertaken in
an attempt to reduce the system losses. Experimental and simulation results of the modifications are presented later in this chapter. The next few subsections discuss some modeling details and validation of the system model.

4.2 Droplet Size Distributions

The DSDs used for the simulations are taken from the literature and discretized into sectional distributions with bin spacings of 0.1 µm. In the case of the AGF 2.0 discussed below, the bins under a diameter of 1 µm have spacings of 0.01 µm. Representative volume fractions for these nebulizers are shown in Figure 4.3.

Kuhli et al. [85] measured the production from a low flow, Aeroneb OnQ nebulizer (VMD = 4.09 µm) using three different methods including optical particle counting, cascade impaction and laser diffraction. The droplet diameters from this distribution are shifted (i.e. increased) in order to model a high flow Aeroneb OnQ nebulizer (VMD = 6.35 µm).

Rotovera et al. [25] measured the output from a Pelas Technology AGF 2.0 Aerosol Generator using laser diffraction. This generator is used in the aerosol shock tube of Texas A&M University. This device employs compressed air to create the droplets and the maximum aerosol concentration is 0.01 mL liquid per L of gas (i.e. $\phi_{\text{dodecane}} \approx 0.04$ in air at 297 K) thus diluted conditions ($O_2 < 21 \%$) must be utilized in the ignition tests.
Figure 4.4: Internal geometries of (a) the initial valve housing [#1], and the second generation housings with entry on the (b) side [#2] and (c) base [#3] of the housing cylinder. The boss on the inlet to the housing accepts the valve stem and the red arrows show the direction of aerosol flow.

While high liquid concentrations are not physically achievable with this device (liquid volume ratio $< 0.01 \text{ mL}_{\text{liq}}/\text{L}_{\text{gas}}$), it is useful to compare its performance to the Aeroneb nebulizers since much smaller droplets are generated and this may be useful for investigating larger hydrocarbon and oxygenated hydrocarbon fuels.

Shadowgraphy and phase doppler anemometry (PDA) techniques were attempted for this thesis as a means of experimentally quantifying the DSD changes experienced through the delivery system and further validating the aerosol model. However, these measurements were not successful. A discussion of these is presented in Appendix E. Primary validation of the aerosol model is achieved through the gravimetric measurements.

4.3 Model Validation and System Modifications

A series of system modifications was undertaken in an attempt to decrease substantial aerosol losses in the system. The modifications as well as measured and predicted losses are discussed in this section. The data for the cyclone mixing chambers was collected by Colin Banyon. Refer to Table 4.2 for the operating conditions used for these experiments.

Aerosol losses in the initial valve housing (#1) of the system were high considering the small amount of residence time in the component. In order to reduce losses, the valve housing was redesigned so that the aerosol is introduced into the cylinder along a wider bend and larger cross-section; a 19.1 mm ID tube with 35° curve of radius of 76.2 mm is
Table 4.2: Operating conditions for modified aerosol system experiments

<table>
<thead>
<tr>
<th>Case</th>
<th>Mixing Chamber Design</th>
<th>Delivery Tube ID (mm)</th>
<th>Delivery Tube Length (m), Position</th>
<th>Valve Housing Design</th>
<th>Hose Barb used to connect DT and VH</th>
<th>Valve present in VH</th>
<th>Water Flow Rate from Nebulizers (g/min)</th>
<th>Gas Flow Rate (L/min)</th>
<th>System Temperature (K)</th>
<th>System Pressure (bar)</th>
<th>Aerosol Concentration (mL/L)</th>
<th>Total System Aerosol Loss (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Box</td>
<td>12.7</td>
<td>1.65, Horizontal</td>
<td>#3</td>
<td>no</td>
<td>no</td>
<td>1.32</td>
<td>18.8</td>
<td>297</td>
<td>1</td>
<td>0.070</td>
<td>14.10±1.92</td>
</tr>
<tr>
<td></td>
<td>Box</td>
<td>12.7</td>
<td>1.65, Horizontal</td>
<td>#3</td>
<td>no</td>
<td>yes</td>
<td>1.34</td>
<td>19.1</td>
<td>297</td>
<td>1</td>
<td>0.070</td>
<td>15.70±1.92</td>
</tr>
<tr>
<td></td>
<td>Box</td>
<td>12.7</td>
<td>1.65, Horizontal</td>
<td>#5</td>
<td>no</td>
<td>no</td>
<td>1.38</td>
<td>32.1</td>
<td>297</td>
<td>1</td>
<td>0.043</td>
<td>7.27±1.44 (w/o RC)</td>
</tr>
<tr>
<td></td>
<td>Box</td>
<td>25.4</td>
<td>1, Horizontal</td>
<td>#5</td>
<td>no</td>
<td>no</td>
<td>1.33</td>
<td>18.8</td>
<td>298</td>
<td>1</td>
<td>0.071</td>
<td>13.64±1.92</td>
</tr>
<tr>
<td></td>
<td>Cyclone</td>
<td>25.4</td>
<td>0.8, Vertical; 0.2, 55° bend</td>
<td>#5</td>
<td>no</td>
<td>no</td>
<td>0.94</td>
<td>19.8</td>
<td>297</td>
<td>1</td>
<td>0.027</td>
<td>7.77±1.92</td>
</tr>
<tr>
<td></td>
<td>Cyclone</td>
<td>25.4</td>
<td>0.8, Vertical; 0.2, 55° bend</td>
<td>#5</td>
<td>no</td>
<td>no</td>
<td>1.09</td>
<td>40.0</td>
<td>297</td>
<td>1</td>
<td>0.027</td>
<td>2.77±1.92</td>
</tr>
<tr>
<td></td>
<td>Cyclone</td>
<td>25.4</td>
<td>0.8, Vertical; 0.2, 55° bend</td>
<td>#5</td>
<td>no</td>
<td>no</td>
<td>2.38</td>
<td>30.1</td>
<td>298</td>
<td>1</td>
<td>0.079</td>
<td>15.04±1.92</td>
</tr>
</tbody>
</table>
used. Two variations of the housing were made where the aerosol is introduced to either the side (#2) or base (#3) of the cylinder. The second generation housings were machined from aluminum (#2) or made from ABS plastic (#3) in a rapid prototyping machine, respectively. The internal geometries of the initial and redesigned valve housings are depicted in Figure 4.4.

The aerosol model predicts that the loss in the second generation valve housing will be reduced to 1.04 % when no valve is present and 1.42 % when a valve is present. Unfortunately however, the model is not able to indicate whether there is a difference between the performance of the side or base entry. Model predictions match fairly well with experiment measurements of aerosol loss for Case 4 & 5 and the comparison is presented in Figure 4.5.

In the initial configuration particles flow into a sharp angle where the aerosol must change direction quickly which allows inertia to carry the larger particles into the wall. By adding the curve, directional changes in the flow field are not as drastic and larger particles have more time to adjust and not deposit on the walls.

The difference between the measured and modeled delivery tube loss is likely a result of the tube position in the experiment. When performing the experiments, the position of the tube was not held constant or recorded. On the other hand, the position of the tube was assumed to be horizontal for the simulations. In the experiment without the valve, the configuration of the tube was likely positioned close to horizontal for the entire
Figure 4.6: Measured and modeled losses in aerosol system components using valve housing #3. Conditions for the system are $T = 297$, $P = 1$ bar, $\dot{m}_l = 1.38$ g/min and $Re_{RC} = 879$.

At a higher flow rate (i.e. $Re_{RC} = 879$) the model predicts that the losses will be reduced in each component as shown in Figure 4.6. This is the case for the experimental measurements of the mixing chamber and valve housing for Case 6, but not for the delivery tube. The loss in the reaction chamber was not measured. The discrepancy in the delivery tube can once again be attributed to the configuration of the delivery tube in the system. In this instance, a portion of the delivery tube may have been positioned vertically, which works to reduce deposition losses. In the figure, it can also be seen that modeled value for the valve housing overpredicts the measured losses. This discrepancy might be due to the fact that the flow through the tube is in the transition regime between laminar and turbulent flow (i.e. $Re_{tu} = 3516$) and the model for removal in the tube bend was not developed for this regime where droplet penetration is generally higher.

The next modification to the system changed the 12.7 mm ID delivery tube to a 25.4 mm ID Tygon tube with a length of 1 m. Two more valve housings with a side (#4) and base (#5) entry to the cylinder were fabricated from ABS using the rapid prototyping machine where the internal passages were increased from 19.1 to 25.4 mm. The model predicts that the loss in the valve housing and delivery tube will remain about the same. These predictions are compared to the experiment losses for Case 7 in Figure 4.7.
Figure 4.7: Measured and modeled losses in aerosol system components using valve housing #5. Conditions for the system are $T = 298$, $P = 1$ bar, $\dot{m}_l = 1.33$ g/min and $Re_{RC} = 516$.

The measured loss decrease seen in the valve housing for Case 7 relative to Case 4 occurs for two reasons. First, the velocity of the flow in the tube is reduced due to the larger diameter. This decreases the stopping distance of the particles. Second, the diameter of the tube is increased. These changes reduce the Stokes number of particles and a particles with lower Stokes number is less likely to be removed from an aerosol. Due to the quasi-empirical sub-model for the bend though, these mechanisms are not completely captured in the model.

Once again, the difference in predicted and measured losses in the delivery tube seen in the data is likely due to the tube position. The 25.4 mm ID tube that was utilized had a gentle arc that could not be straightened. As a result the tube had both inclined and declined portions along its length which result in less deposition than would be expected with a horizontal tube. Predicted losses in the delivery tube remain the same because the effects of increasing the diameter and decreasing the length oppose each other. The larger diameter decreases the flow though the tube. This means the aerosol remains in the tube longer and particles are more likely to settle out. On the other hand, shortening the tube decreases the retention time and less particles are lost for the same reason.

Another modification made to the fuel delivery system was to change the mixing chamber from the box geometry to a cyclone with a 0.152 m diameter $\times$ 0.254 m high cylinder and a cone with a half angle of $\Theta = 35^\circ$. The bottom of the mixing chamber opens to the 1 m $\times$ 25.4 mm ID delivery tube which is approximately positioned such that
Figure 4.8: Delivery system with cyclone mixing chamber. The delivery tube is not to scale.

a 0.8 m vertical section is followed by a 55 degree curve at a radius of 0.2 m before meeting with the valve housing. A schematic of the system is presented in Figure 4.8. The new configuration was designed to have well controlled mixing of droplets and gas.

Results from the model and experiment are shown in Figure 4.9. The model predicts that the losses in the mixing chamber will be about the same as the box mixing chamber and losses in the delivery tube will be smaller at similar conditions. The measured results are in agreement with the delivery tube, but the mixing chamber losses are less than half of the predicted values. The model does however predict the correct trend where losses decrease as the Reynolds number of the reaction chamber increases. The discrepancy in the magnitude of the loss could possibly be attributed to the aerosol not sweeping the entire volume of the mixing chamber. This might happen if the aerosol only flows along the outside of the cylinder with a dead zone in the center of the cylinder.

The possibility of incomplete sweeping of the mixing chamber was investigated for Case 8 to determine the effect on the losses of the other components of the system. The model results in Figure 4.10 show that the level of losses in the mixing chamber has a
Figure 4.9: Measured and modeled losses in aerosol system components using valve housing #5 and the 0.152 m diameter cyclone mixing chamber. Conditions for the system are $T = 297$, $P = 1$ bar. For $Re_{RC} = 543$, $\dot{m}_l = 0.94$ g/min and for $Re_{RC} = 1094$, $\dot{m}_l = 1.09$ g/min.

Figure 4.10: Modeled losses in aerosol system components for a full and half sweep of the mixing chamber using valve housing #5 and the 0.152 m diameter cyclone mixing chamber. Conditions for the system are $T = 297$, $P = 1$ bar, $Re_{RC} = 543$ and $\dot{m}_l = 0.94$ g/min.
minimal effect on the losses of components down system from the mixing chamber. The same pair of simulations was conducted for lower gas flow, higher liquid flow aerosols with similar results. A more rigorous model is necessary to ascertain the reason for the discrepancy, but the full and half sweep simulations show that even with uncertainty in the mixing chamber losses, losses in the other components can be reasonably predicted using the model.

For the delivery tube, losses are decreased because the majority of the tube is vertical, therefore there is no surface for settling droplets to collect until the tube bend.

The final change to the delivery system was to increase the cyclone diameter to 0.205 m and was designed to accommodate up to 10 nebulizers in order to prepare fuel rich (i.e. $\phi > 1.0$) aerosols of high concentration. For this geometry though, the model predicts that the loss in the mixing chamber will increase significantly. The simulations also indicate that the loss in the delivery tube is expected to decrease while the loss in the reaction chamber should increase, at least for the typical conditions expected to be utilized. The model results are compared to experimental measurements in Figure 4.11.

In the experiment data and the model for Case 10, it should be noted that the DSD from the nebulizers is thought to be slightly larger. This is probably due to problems with the nebulizer manufacturer where adequate control could not be achieved with the nebulizers provided to the laboratory. The overall output for these nebulizers is approximately 0.60 g/min compared to 0.33 g/min for the lower VMD nebulizers.
The trend in the model and experiments can be explained as follows. The volume of the 0.205 m diameter cyclone is \(\approx 1.7\) times larger than the other mixing chambers and therefore the retention time is increased leading to larger losses. The larger droplet sizes also contribute to the increase in mixing chamber loss because they have larger settling velocities. The increased deposition in the reaction chamber is due to the larger initial droplets and the fact that fewer droplet are removed in the delivery tube than previous configurations.

It can be seen in the validation experiments that the aerosol model provides reasonably accurate predictions of the expected losses in most fuel delivery system components as the system configuration and operating conditions are changed. Another method of validation not pursued in this study would be to measure the temperature at the mixing chamber inlet and outlet. Comparing these measurements to model results would help to confirm the evaporation and heat transfer models used.

Comparisons of experiment and model results show that the mechanisms in the mixing chamber cannot be completely captured by the bulk model, however, the level of losses in the mixing chamber do not significantly impact the level of losses in other system components. In the next section the model is used to understand the important mechanisms that effect the aerosol, follow the aerosol behavior inside the system, and investigating conditions that could not be tested using the experimental setup.

### 4.4 Model Utilization

In this section, the model is used to understand the behavior of the aerosol in the delivery system and predict how the system should behave over a range of parameters. The effect of mechanisms, conditions along the system, operating maps of the aerosol system and potential system improvements are presented next.

Unless otherwise stated, the simulated aerosol system is composed of:

1. Cyclone mixing chamber (.205 m ID, 35° tapered cone)

2. Delivery tube (25.4 mm ID) with a 50.8 mm vertical section followed by a 55° bend
Figure 4.12: Percent loss in delivery systems components for \( n \)-dodecane in air with (a) \( \phi = 1.0 \) and (b) \( \phi = 2.0 \) at \( T = 350 \) K, \( P = 1 \) bar and \( \text{Re}_{RC} = 800 \).

with a 76.2 mm radius

3. 2nd generation valve housing (#5) with a 25.4 mm ID inlet and poppet valve (6.1 mm OD stem, 31 mm OD seat)

4. Reaction chamber (51 mm ID, 0.305 m length)

4.4.1 Effect of Mechanisms

Simulations of \( n \)-dodecane in air (21 % \( \text{O}_2 \), 79 % \( \text{N}_2 \)) are conducted with sub-models for coagulation and evaporation sequentially turned on and off in order to ascertain the effect on aerosol loss. The simulation conditions and results for these tests are presented in Figure 4.12. A reaction chamber Reynolds number of 800 is used (40.1 LPM gas) with \( T = 350 \) K and \( P = 1 \) bar. Fuel loadings of \( \phi = 1 \) and 2 are investigated.

At low fuel concentrations (i.e. \( \phi = 1 \)), it can be seen that evaporation has a major effect on the losses in an aerosol system because a large portion of the liquid mass is vaporized. More droplets completely evaporate, lowering the particle concentration, and the remaining droplets are reduced in size. On the other hand, because of the low concentration conditions present coagulation has a minimal effect. Results when the sub-model for Brownian coagulation is turned off are nearly identical to the results for all models turned on, which is expected since Brownian coagulation is only significant for particles below 1 \( \mu \)m. Results when the sub-model for gravitational coagulation is turned
4.4.2 Evolution of DSD

The behavior of the aerosol DSD is also of interest since the droplets will need to be evaporated during the compression stroke of the RCM’s pistons. In particular it is the largest droplets that will require the longest time to evaporate, so these must be as small as possible. The volume fraction DSD at the exit of each component in the delivery system is therefore tracked for each of the conditions used in Section 4.4.1. In both cases, it is found that the DSD is modified substantially only during the time that the aerosol is retained in the mixing chamber. Thereafter, the DSD remains constant until it exits the system.

A plot showing the initial and final DSDs is provided in Figure 4.13. The volume fraction contained in the smaller droplets is first reduced as the droplets vaporize. Once
the gas phase is saturated, the smaller particles coagulate to form larger particles. This moves mass to the larger particles as is shown in the plot. Only a slight difference is present between the final DSDs for the two equivalence ratios and in particularly the peak droplet size is unaffected by the fuel concentration of these conditions. As a percentage, more mass exists in the gas phase for the lower equivalence ratio. This mass comes mainly from the smaller droplets as they evaporate more quickly. The end result is a lower percentage of small particles and greater percentage of large particles when compared to the higher equivalence ratio.

### 4.4.3 Aerosol Behavior

The aerosol model is next used to track key parameters of the aerosol as it is transported through the delivery system; these include the relative humidity (RH), mass percent evaporated (MPE), mass percent lost (MPL) and aerosol temperature. The parameters are defined as

\[
\text{RH} = 100 \times \frac{P_{\infty}}{P_{\text{sat}}} \quad (4.2)
\]

\[
\text{MPE} = 100 \times \frac{m_v}{m_{l,0}} \quad (4.3)
\]

\[
\text{MPL} = 100 \times \frac{m_{l,0} - m_l - m_v}{m_{l,0}} \quad (4.4)
\]

For the investigation Case 1 is simulated with a flow rate of 18.8 LPM at \( T = 298 \) K, \( P = 1 \) bar (\( \text{Re}_{RC} = 516 \)) for the inlet gas. A liquid concentration at 0.070 mL/L with water and \( n \)-dodecane as the liquids. Results from these simulations are shown in Figure 4.14. Here it can be seen that initial evaporation occurs very quickly (\( \approx 100 \text{ ms} \)) with a corresponding drop in temperature. As the aerosol moves through the system its temperature, MPE and MPL slowly increase. When the aerosol reaches the delivery tube a sharp rise in temperature occurs due to the increased heat transfer as the aerosol is more exposed to the system boundaries. The MPE increases due to the increased temperature while the MPL increases due to the more constricting geometry. The same trends continue through the valve housing. As the aerosol moves into the reaction chamber, the temperature and MPE begin to stabilize as contact with the system
Figure 4.14: Relative humidity, mass loss, mass evaporation and aerosol temperature along the delivery system for (a) water and (b) n-dodecane at $T = 298$ K and $P = 1$ bar. The time in the mixing chamber (MC), delivery tube (DT) and reaction chamber (RC) is marked. The time in the valve housing is 0.01 s.

boundaries is decreased. A slight increase in the MPL occurs as the aerosol moves through the reaction chamber and exits the system.

The RH for each liquid reaches 100 % after the initial evaporation and remains at this level through-out the system. The unsteadiness in the water is attributed to the fact that parameters in the model are optimized for n-dodecane. Using smaller evaporation steps would result in an improved trace for the RH of water, as discussed in Appendix B.

The difference in MPE is due to the difference in saturation pressures of the two liquids. Water has a saturation pressure of about 0.03 bar at 297 K while $P_{\text{sat}}$ is 0.00014 bar for n-dodecane. While the vapor pressure of n-dodecane is 0.5 % of the value for water, the MPE of n-dodecane is 5 % of the value for water. This factor of 10 is because of the difference in molar mass between the two liquids: 18.02 and 170.3 g/mol for water and n-dodecane, respectively.

The MPL for the water and n-dodecane are similar, with the values for n-dodecane being smaller than values for water. This result is due to the differing densities of the two liquids. The density of n-dodecane is 75 % of the value for water. This means that a particle of n-dodecane will have a smaller terminal settling velocity and a lower Stokes number than an equal sized particle of water.

The difference in the scale of the temperature traces for the two liquids is due to
Figure 4.15: Relative humidity, mass loss, mass evaporation and aerosol temperature along the delivery system for \( n \)-dodecane at \( T = 350 \) K and \( P = 1 \) bar. The time in the mixing chamber (MC), delivery tube (DT) and reaction chamber (RC) is marked. The time in the valve housing is 0.01 s.

the different vaporization enthalpies. Water has \( h_{fg} \approx 2,440 \) kJ/kg while that for \( n \)-dodecane is \( \approx 350 \) kJ/kg, so that the bath gas experiences a larger temperature drop with water.

A simulation was also run for \( n \)-dodecane in the cyclone fuel delivery system described in Section 4.3 with \( T = 350 \) K, \( P = 1 \) bar, \( \phi = 1.0 \) and \( Re_{RC} = 800 \). The results are shown in Figure 4.15 and differ from the simulation at lower temperature. When the aerosol reaches the reaction chamber 44.5% of the mass is in the vapor phase. This is more than water at 298 K and the reason is because of the difference in molar mass, as discussed previously. With such a large percent of mass in the vapor phase, the remaining aerosol droplets are smaller than at low temperature and thus they are not lost as easily in the delivery system.

4.4.4 Operating Maps

The aerosol model was used to generate operating maps for the eight different aerosol systems listed in Table 4.3. Simulations were run for overall fuel loading equivalence ratios up to 2.0 with reaction chamber Reynolds numbers between 200 and 1200 as these are the conditions under which RCM experiments are expected to be performed. Equivalence
Table 4.3: Operating conditions for simulated aerosol systems.

<table>
<thead>
<tr>
<th>System</th>
<th>1</th>
<th>2</th>
<th>3</th>
<th>4</th>
<th>5</th>
<th>6</th>
<th>7</th>
<th>8</th>
</tr>
</thead>
<tbody>
<tr>
<td>Nebulizer</td>
<td>OnQ</td>
<td>OnQ</td>
<td>OnQ</td>
<td>OnQ</td>
<td>AGF 2.0</td>
<td>AGF 2.0</td>
<td>OnQ</td>
<td>OnQ</td>
</tr>
<tr>
<td>Gas</td>
<td>Air</td>
<td>Air</td>
<td>32% Ar, 47% N₂, 21% O₂</td>
<td>36% Ar, 53% N₂, 11% O₂</td>
<td>Air</td>
<td>Air</td>
<td>Air</td>
<td>Air</td>
</tr>
<tr>
<td>Fuel</td>
<td>C₁₂H₂₆</td>
<td>C₁₂H₂₆</td>
<td>C₁₂H₂₆</td>
<td>C₁₂H₂₆</td>
<td>C₁₂H₂₆</td>
<td>C₁₆H₃₄</td>
<td>C₁₂H₂₆</td>
<td>C₁₆H₃₄</td>
</tr>
<tr>
<td>T₀ (K)</td>
<td>300</td>
<td>350</td>
<td>350</td>
<td>350</td>
<td>350</td>
<td>350</td>
<td>350</td>
<td>350</td>
</tr>
<tr>
<td>P₀ (bar)</td>
<td>1</td>
<td>1</td>
<td>1</td>
<td>1</td>
<td>1</td>
<td>1</td>
<td>0.5</td>
<td>0.5</td>
</tr>
</tbody>
</table>

Figure 4.16: Operating map of System 1: Air + C₁₂H₂₆, OnQ nebulizer, T = 300 K, P = 1 bar. The range of conditions for the experiments with water is indicated.

The overall system loss and the droplet diameter under which 98% of the ‘fuel’ mass is contained, d₉₈, in the reaction chamber are predicted for each of the operating conditions. These values indicate the potential for performing an experiment in an aerosol RCM. High losses are not acceptable because they will lead to uncertainty of fuel loading, more frequent cleaning of equipment, and contamination of experiment results. Small droplets (i.e. < 4 μm for n-dodecane) are desired to ensure complete evaporation and mixing during the compression of the RCM piston.

The predicted performance of System 1 is shown in Figure 4.16 where the stratification limit for the OnQ nebulizer, determined by Johnson [28], is indicated. For lower Reynolds numbers, the loss in the aerosol system is almost entirely dependent on...
the overall fuel loading. As the flow rate increases, loss in the aerosol system decreases as expected. However, the predicted losses for the highest flow rate are still unacceptable for RCM experiments. The reduction in losses is mainly due to the decreased retention time in the delivery system. Reducing the time that the aerosol is contained in the delivery system limits the number of droplets that settle out of suspension.

Figure 4.16 also indicates the initial $d_{98}$ value, $d_{98,0}$, for the OnQ nebulizer of 11.7 $\mu$m. This value will decrease as droplets evaporate and the larger particles fall out of suspension. At the same time, particles will also coagulate and increase the $d_{98}$ value. The $d_{98,0}$ line indicated on the graph shows where these mechanisms are balanced. Above the line, coagulation is more of a factor, and below the line, evaporation and removal are larger factors.

In Figure 4.16 the mechanisms are balanced at an equivalence ratio of $\approx 0.5$. When the fuel loading is above 0.5, coagulation is the dominant mechanism affecting the DSD. It is also important to note the spacing between the $d_{98}$ contour lines. For slower flows, droplet size increase is more pronounced as the fuel loading increases. This is once again due to the increased retention time in the aerosol system, which allows more time for the mechanisms to affect the DSD.

Another important feature of the plot in Figure 4.16 is that the vast majority of fuel is in the liquid phase for the 'low' temperature system. At 300 K, the gas phase has a saturated equivalence ratio of 0.016. The large amount of fuel in the liquid phase results in the performance behavior discussed above. With little evaporation, small droplets do not disappear and large droplets do not decrease in size.

The behavior of the other systems listed in Table 4.3 are discussed next. For System 2, the temperature is increased to 350 K. The operating map presented in Figure 4.17 shows that the increased temperature leads to significant decreases in aerosol losses. Also shown is that the $d_{98}$ values are lower than System 1 for equivalent operating conditions. In this system more of the fuel evaporates because the higher temperature increases the vapor pressure of the fuel. Thus, when the global equivalence ratio is below $\approx 0.5$ all of the fuel is in the gas phase.
The increased evaporation is the main reason for both of the changes seen in the operating map of System 2. Evaporation causes smaller droplets to completely vaporize thereby reducing the aerosol droplet count density and also causes larger droplets to decrease in size. Smaller particles are less affected by gravity and remain in suspension. Smaller particles in aerosols with lower droplet count densities are also less likely to coagulate and therefore the $d_{98}$ values are not as large.

In both systems, the $d_{98}$ value increases with increased fuel loading and, as stated earlier, losses decrease as the flow rate increases. However, when the equivalence ratio is near the level where the fuel is completely in the gas phase, the dependence on flow rate is not as strong. This is when the droplets are smallest and do not settle out of the aerosol.

In System 3, the gas is composed 32 % Ar, 47 % N$_2$ and 21 % O$_2$ by volume. This represents a test gas that is prepared using 21 % O$_2$ and 79 % of a 60/40 blend of N$_2$/Ar. Argon is commonly used in RCM experiments due to its low heat capacity which allows for increased compression temperatures to be achieved relative to using air at the same compression ratio. Argon mixtures extend the temperature regime where an RCM can operate. However, the larger molar mass of argon affects its transport properties and this would impact performance of the aerosol fueling system.

The operating map for System 3 is shown in Figure 4.18. Here it is seen that the
Figure 4.18: Operating map of System 3: 32 % Ar / 47 % N\textsubscript{2} / 21 % O\textsubscript{2} + C\textsubscript{12}H\textsubscript{26}, OnQ nebulizer, $T = 350$ K, $P = 1$ bar.

map is almost identical to the map of System 2 with the $d_{08}$ and loss values slightly larger for the individual operating conditions. This indicates that there is not a significant change in behavior of aerosol particles between the air and the Ar/N\textsubscript{2}/O\textsubscript{2} gas mixture, even though the Ar mixture has a higher molar density which can reduce evaporation rates and increase rates of heat transfer, as well as wall friction.

System 4 is configured using a gas mixture that represents a gas composed of 36 % Ar, 53 % N\textsubscript{2} and 11 % O\textsubscript{2}. Dilute mixtures are often used in STs and RCMs in order to minimize extreme pressures after ignition takes place as well as decrease the chemical reactivity in order to prevent reaction during the piston compression. The operating map for this system is shown in Figure 4.19, and indicates different behavior from the previous system. In this case, the mass of fuel needed to obtain a particular equivalence ratio is less than the previous systems because of the reduced O\textsubscript{2} concentration. At the same time, the mass of fuel that enters the gas phase is the same as in the previous system. This leads to a gas phase with a higher achievable equivalence ratio and a liquid phase that generally has smaller droplets.

In Figure 4.19, it can be seen that once the system operates above the stratification limit, increasing the fuel loading only has a small effect on the $d_{08}$ value and the flow rate has virtually no effect. The contour for the $d_{08,0}$ value is no longer present on
Figure 4.19: Operating map of System 4: 36 % Ar / 53 % N₂ / 11 % O₂ + C_{12}H_{26}, OnQ nebulizer, T = 350 K, P = 1 bar.

the plot indicating that the large extent of evaporation in this system reduces the droplet sizes below this value.

System 5 used the same parameters as System 2 with the AGF 2.0 droplet distribution, as opposed to the OnQ droplet distribution. The results in Figure 4.20 show that a significant improvement in performance can be made by using smaller droplets. While the trend of the aerosol loss contour lines is similar to previous systems, the amount of loss is at the level where it will have almost no effect on an actual RCM experiment. The stratification limit for the AGF 2.0 nebulizer has not been investigated unfortunately and is expected to be significantly below the range presented on the operating map.

Comparing the operating maps for System 2 and 5 indicates that the droplet size does not impact the amount of fuel that evaporates in the fueling system. This means that the time scale for evaporation is much smaller than that for transport of the aerosol through the delivery system. The plots also show that coagulation has only a limited effect on the smaller droplets. In System 2, the range of $d_{98}$ values is smaller for the high flow rates. This is because the retention time in the system is decreased and there is less time for particles to coagulate. In System 5, this trend is not present and the $d_{98}$ value is independent of the flow rate.

System 6 uses the same conditions as System 5, however the fuel is $n$-hexadecane,
Figure 4.20: Operating map of System 5: Air + C_{12}H_{26}, AGF 2.0 nebulizer, $T = 350$ K, $P = 1$ bar.

Figure 4.21: Operating map of System 6: Air + C_{16}H_{34}, AGF 2.0 nebulizer, $T = 350$ K, $P = 1$ bar.
as opposed to \textit{n}-dodecane. \textit{n}-Hexadecane is a larger fuel molecule (i.e. 16 carbons as opposed to 12) and is more representative of biodiesel than \textit{n}-dodecane. The operating map for this system is shown in Figure 4.21.

Compared to the previous system, the equivalence ratio of the vapor phase is much smaller because the vapor pressure for \textit{n}-hexadecane is much lower than \textit{n}-dodecane. Also, once the equivalence ratio is above 0.25, the aerosol loss is predicted to be dependent only on the flow rate. This change in behavior can also be attributed to the saturation pressure since the other thermophysical properties of the two fuels are very similar.

The parameters for System 7 are similar to System 2, however the system pressure is reduced from 1 bar to 0.5 bar. This condition will reduce the peak compressed temperature during the experiment as well as reduce the chemical reactivity and alter the fuel’s evaporation characteristics. The results from the simulation with this system are shown in Figure 4.22. As it can be seen, the equivalence ratio supported by the gas phase is once again increased because the lower amount of \textit{O}_2 in the bath gas.

The losses decrease significantly compared to System 4 where the \textit{O}_2 level is about the same. The reduction in loss seen here is a result of the increased flow. In order to maintain the same Reynolds number at 0.5 bar, the flow rate must be doubled because the kinematic viscosity of the gas is doubled. Increased flow rate results in less residence time
The plot shows that an aerosol in this system is not retained long enough to be affected by coagulation. On the other hand, the simulation results for this case indicate that the Reynolds number based on the bulk scale characteristics might not be the best value for comparison between all systems. The changing pressure has a direct impact on the Reynolds number, but does not significantly alter the Stokes number of the droplets, which is a more important parameter affecting the individual droplet behavior.

In the final System, \( n \)-hexadecane is used as the fuel under the same parameters as in System 7. The results are shown in Figure 4.23 and are once again much different than the system using \( n \)-dodecane. The reduced effect of evaporation due to the low saturation pressure of the fuel leads to a system where losses are only dependent on the gas flow rate. However, once the flow rate is above the stratification limit, the effect of the flow rate on the losses is greatly reduced and the range of \( d_{98} \) values is very small. Both of these results can be attributed to the reduced retention time.

It can be seen that the trends for losses in the reaction chamber follow closely with the trends in the overall loss in the aerosol system. For the OnQ nebulizer droplet distribution, the losses in the reaction chamber generally account for 21% of the overall system loss. The most significant contribution to the aerosol loss comes from the mixing
chamber, which on average, makes up about 65 % of the overall system loss, while the
curved portion of the delivery tube contributes the remaining losses. On the other hand,
for the AGF 2.0 nebulizer, the reaction and mixing chambers contribute roughly 13 and
80 % of the overall loss, respectively.

4.5 Fuel Delivery System Improvements

Based on the results from simulations conducted in the previous section, it appears that
the most effective way to improve the system performance is to generate aerosols with
very small droplets. Unfortunately, current technologies capable of generating small
droplets are not able to achieve high enough fueling loads. Even though this is the case,
the simulations do provide insight into the droplet distributions that are needed in order
to deliver an appropriate droplet size to the reaction chamber. At 350 K and above a
reaction chamber Reynolds number of 500, the delivered distributions had a $d_{98}$ that was
no larger than 7 % of the original value and the majority of operating conditions had $d_{98}$
values that were lower than the initial value. Therefore, a delivered droplet distribution
should have a $d_{98}$ that is equal to the desired delivered value.

Simulations and experiments also showed that systems with larger droplets (i.e. >
1 $\mu$m) performed best when the retention time is the smallest, because the effects of
coagulation and removal are reduced. Therefore, each component in the system needs to
be properly sized. This is most important for the mixing chamber, where the majority of
system losses occur. Properly sized for this component means that once the aerosol is
homogeneous it should enter the delivery tube.

Another design feature that could be modified to further reduce losses in the
system would be to remove the curved portion of the system (i.e. delivery tube and valve
housing). This would require the use of a different type of valve (e.g. ball, gate, or
butterfly) that would need to be designed for the high temperature and pressure
environment experienced in an RCM. The poppet valve was originally used because of its
simplicity and low cost. Removing the curve also means that the mixing chamber outlet
would need to be on the side similar to the box-type mixing chamber.
4.6 Areas for Model Improvement

The major area for improvement in the aerosol model lies in the bend removal sub-model. A semi-empirical fit of the data was used and is only shown to correlate to experimental results at one geometry of the tube bend. Therefore, the model is not able to test the effect of changing the geometry of a bend in the system.

Another area for improvement is the mixing chamber removal sub-model. The bulk model presented is not able to predict the magnitude of aerosol loss. It assumes that all particles move through the system at the same rate and this is not the case. Larger particles move to the exit of the mixing chamber more quickly than the smaller molecules because of increased settling velocity. In order to more accurately predict the losses, a more refined method that can track particle location is necessary.

Finally, multiple assumptions were made in the model in order to simplify the calculations and allow for exploration of the system performance over a range of operating conditions and various system setups. Therefore, it would be useful to validate the assumptions by comparing model results to other studies of the aerosol system. CFD simulations or measurement of droplet distributions along the aerosol system are two potential methods that could be used for additional, more rigorous validation of the system model.
Chapter 5

SUMMARY

The main goal of this thesis was to develop a model that could be used to understand the behavior of dense aerosols in a fuel delivery system under laminar flow conditions, as well as predict system performance over a range of operating conditions. This study is applicable the development of an aerosol RCM that can be used to measure the chemical kinetics of low-volatility fuels such as biodiesel.

The model predictions for aerosol losses in a fuel delivery system mock-up matched fairly well with measured results from experiments. The experiments were conducted with an aerosol of water in air at $T = 297$ K, $P = 1$ bar and at system flow rates of 19 and 33 liters per minute. The loss in the mixing chamber could not be consistently predicted, however, the trends in performance were captured reasonably well. It was shown that the magnitude of the predicted mixing chamber loss did not have a large effect on the level of losses in other components and, therefore, the model could be used to predict losses in the reaction chamber. Any loss in the reaction chamber can affect the RCM experiment results and it is important to know what levels of loss are expected.

The model was used to gain an understanding of important mechanisms and to predict system performance of fuel aerosols in various fueling systems and operating conditions. The study showed that the generated and delivered DSDs do not differ greatly, and for most operating conditions, the delivered DSD has a smaller $d_{98}$ value than the the generated DSD. This is an important result to consider for the design of the delivery system. Initially, it was suspected that smaller droplets would lead to increased coagulation due to the high particles densities, but this was not shown to be a problem.

The study also showed that gravity is the most important force influencing the
behavior of the aerosols in the system. It is responsible for large droplets settling out of suspension and coagulating with smaller, slower settling droplets before depositing on system components. Therefore, it both causes and enhances the loss in the system.

The simulations show that one technique to reduce the effect of gravity is to increase the flow rate through the delivery system. This decreases the time over which gravity can impact an aerosol. Another way to reduce the effect of gravity is to perform experiments with smaller aerosol droplets. However, generating droplets that are small enough in a high enough concentration aerosol is a major challenge that would need to be overcome in order to successfully perform an actual RCM experiment. Finally, another method, though probably not as practical, would be to reduce the gravitational force itself and conduct experiments in microgravity conditions.
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Appendix A

THERMOPHYSICAL PROPERTY MODELS

All models with the exception of gas enthalpies are used as recommended by Poling, et al. [75].

Gas Density

The Ideal Gas equation of state was used to calculate the density of gases in the model.

\[ \rho_g = \frac{PM_g}{R_u T} \]  \hspace{1cm} (A.1)

Gas Viscosity

The model used to calculate the dynamic viscosity of a gas suggested by Lucas [86] is

\[ \mu_g = \frac{1}{\zeta} \left[ 0.807T_r^{0.618} - 0.357e^{-0.449T_r} + 0.340e^{-4.058T_r} + 0.018 \right] \]  \hspace{1cm} (A.2)

\[ \zeta = 3.79 \times 10^{10} \left( \frac{T_c}{M_g P_c} \right)^{1/6} \]  \hspace{1cm} (A.3)

\[ T_r = \frac{T}{T_c} \]  \hspace{1cm} (A.4)

Kinematic viscosity is defined as

\[ \nu_g = \frac{\mu_g}{\rho_g} \]  \hspace{1cm} (A.5)
Gas Conductivity

The thermal conductivity of a gas is calculated using the method of Chung, et al. [87] where

\begin{equation}
  k = \frac{3.75 \times 10^{-3} \psi R_a \mu_g}{M_g} \tag{A.6}
\end{equation}

\begin{equation}
  \psi = 1 + \alpha \left( \frac{0.215 + 0.28288 \alpha - 1.061 \beta + 0.26665 Z}{0.6366 + \beta Z + 1.061 \alpha \beta} \right) \tag{A.7}
\end{equation}

\begin{equation}
  \alpha = \frac{C_v}{R_a} - \frac{3}{2} \tag{A.8}
\end{equation}

\begin{equation}
  \beta = 0.7862 - 0.7109 \omega + 1.3168 \omega^2 \tag{A.9}
\end{equation}

\begin{equation}
  Z = 2.0 + 10.5 T_r^2 \tag{A.10}
\end{equation}

\begin{equation}
  C_v = C_p - R_a \tag{A.11}
\end{equation}

Gas Enthalpy and Specific Heat

The enthalpy and specific heat of individual gases are calculated from the polynomial fit with the coefficients coming from the Burcat database [88]. The values for mixtures are computed using a simple molar weighting.

Vapor Diffusivity

The model used to calculate the diffusion coefficient was developed by Fuller, et al. [89].

\begin{equation}
  D_v = \frac{0.0143 T^{1.75}}{P M_{AB}^{0.5} (\Sigma_A^{1/3} + \Sigma_B^{1/3})^2} \tag{A.12}
\end{equation}

\begin{equation}
  M_{AB} = \frac{2}{1/M_A + 1/M_B} \tag{A.13}
\end{equation}

where \( \Sigma_A \) is the summation of atomic diffusion volumes for species A. Representative diffusion volumes for \( H_2O, \) \( n \)-dodecane, \( n \)-hexadecane, Air, \( N_2 \) and Ar are 13.1, 250.9, 332.9, 19.7, 18.5 and 16.2, respectively [75].
Vapor Saturation Pressure

The saturation pressure is calculated using the following equation proposed by Antoine [90]

$$\log_{10}(p_{\text{sat}}) = 5 \left( A - \frac{B}{T + C - 273.15} \right)$$  \hspace{1cm} (A.14)

where the values for A, B and C are tabulated in Poling, et al. [75]. The values for water are A = 5.11564, B = 1687.5370 and C = 230.17. The values for n-dodecane are A = 4.12285, B = 1639.27 and C = 181.84. The values for n-hexadecane are A = 4.15357, B = 1830.51 and C = 154.45. The value for $p_{\text{sat}}$ is given in Pascals.

Liquid Density

The density of a liquid is calculated using the equation proposed by Rackett [91]

$$\rho_p = \frac{\rho_p^R}{\left(0.29056 - 0.08775\omega\right)^\phi}$$  \hspace{1cm} (A.15)

where $\omega$ is the accentric factor and

$$\phi = \left(1 - \frac{T}{T_c}\right)^{2/7} - \left(1 - \frac{T^R}{T_c}\right)^{2/7}$$  \hspace{1cm} (A.16)

where $\rho_p^R$ is the density at reference temperature $T^R$.

Liquid Enthalpy

The enthalpy for liquids, with the exception of water, is determined using the method of Růžička and Domalski [92]. The heat capacity is integrated from 298 K to the liquid temperature and added to the heat of formation at 298 K to determine the enthalpy value. The heat capacity is calculated by

$$\frac{C_p}{R} = \left[ A + B \frac{T}{100} + D \left( \frac{T}{100} \right)^2 \right]$$  \hspace{1cm} (A.17)

where R is the gas constant and A, B and D are parameters obtained using group additivity. The values for n-dodecane are A = 35.6624, B = -1.22961 and C = 1.45768
respectively.

The enthalpy for liquid phase water is calculated using the polynomial fit from the Burcat database [88].

**Thermophysical Property Model Parameters**

A matrix of the parameter values used in the various thermophysical property models is created for each gas mixture and fuel. The matrices are loaded into the main aerosol code in order to compute the properties during the system simulations. Parameter values are gathered from various sources [1, 75, 88, 93, 94].
Appendix B

ALGORITHM VALIDATION

This appendix reviews the validation of various sub-models used in the aerosol fuel delivery model, coefficients for evaporation, and time steps selection for use in the aerosol model. The methods utilized to account for coagulation and evaporation are compared to analytical solutions for monodisperse and bidisperse droplet populations to evaluate issues of numerical diffusion. Next, the value for an appropriate evaporation coefficient is determined using simulations of initial liquid evaporation in the mixing chamber. Finally, the minimum number of coagulation time steps needed in the delivery system components is evaluated.

The analytical solutions used for model validation are derived by Hinds [1] and in some cases an adjustment made by Seinfeld and Pandis [2] is used to modify the solution.

Coagulation

Equation (3.26) is used to model a monodisperse aerosol population with $d_0 = 2 \ \mu m$ and $N_0 = 2 \times 10^{13}$ particles/m$^3$ undergoing coagulation with a constant coefficient of $5.90 \times 10^{-16} \ m^3/s$. The total number of particles is allowed to decrease by one percent during each time step and the model is run for a simulated time of 30 s. Simulations use bin spacings of 0.1, 0.5 and 2.0 $\mu m$ with the largest bin having a size of 40 $\mu m$.

The analytical solution for particle concentration as a function of time is represented as

$$N(t) = \frac{N_0}{1 + 0.5N_0Kt} \quad (B.1)$$

Results of this comparison can be seen in Figure B.1 where water is used as the dispersed
A comparison between the analytical and numerical solution is also made using a bidisperse aerosol with an initial population of $d_{0,1} = 2 \, \mu m$, $d_{0,2} = 10 \, \mu m$ and $N_{0,1} = N_{0,2} = 1 \times 10^{13}$ particles/m$^3$. All other variables are the same as the monodisperse simulation.

The analytical solution for a droplet diameter as a function of time is

$$d(t) = d_0(1 + 0.5N_0Kt)^{1/3} \quad (B.2)$$

The analytical solution is compared to the diameter of average mass for the numerical
solution which is calculated by
\[
d_{\bar{m}} = \left( \frac{\sum n d^3}{N} \right)^{1/3}
\] (B.3)

Results of this comparison are shown in Figure B.2 and once again it can be seen that these are nearly identical.

These simulations indicate that the coagulation algorithm can be used at these conditions without generating any numerical diffusion. One problem that may arise however is if there are not enough bins to capture the majority of the droplets generated. For example, a sectional model has bins that range from 1 arbitrary units (au) to 10 au. If two particles with 6 au coagulate then there is no bin for a 12 au particle to be placed in these simulations. This mass will be lost from the system thereby creating diffusion.

**Evaporation**

Equation (3.15), with Eq. (3.18) as the rate equation, is used to model evaporation of a single water droplet with \( d_0 = 2 \ \mu m, T = 300 \ \text{K} \) and \( P = 1 \ \text{bar} \) where the gas phase is air. Heat transfer is not incorporated into these simulations, but the temperature is assumed to remain constant. The particle is allowed to evaporate so that it moves one bin per time step. The total computed evaporation time is 1.25 ms and simulations are conducted using bin sizes of 0.1, 0.5, and 2.0 \( \mu m \). The sectional method using a range of bin sizes is
Figure B.4: Non-dimensional surface area plotted as a function of time for a monodisperse water droplet population with $d_0 = 2 \mu m$, $N_0 = 1 \times 10^{13}$ particles/kg, $P_{v,0} = 0$, $T = 293$ K and $P = 1$ bar.

compared to the analytical equation and the results are presented in Figure B.3. In this figure the non-dimensional surface area is plotted as a function of time. Typical $d^2$-law behavior is expected for this isolated droplet.

Bin spacings of 0.1 and 0.5 $\mu m$ provide results that closely match the analytical rate equation. Deviation from the rate equation for the 2.0 $\mu m$ spacing is due to droplets moving through the bins too quickly because there is not enough resolution. For example, with bin spacing of 0.1 $\mu m$, the 2.0 $\mu m$ droplet needs to move through 19 bins before evaporation. On the other hand, with bin spacing of 2 $\mu m$, the droplet moves through no bins before evaporation. Droplets either exist or they do not, and this causes droplets to evaporate faster than the analytical model would predict otherwise.

A monodisperse aerosol population with $d_0 = 2 \mu m$, $N_0 = 1 \times 10^{13}$ particles/kg and $P_{v,0} = 0$ is also simulated. In this case the aerosol is confined so that the vapor pressure provided by the evaporating liquid eventually saturates the gas phase. The time step is set so that 10% of the difference between the saturation pressure and vapor pressure was allowed to evaporate each step. In the sectional model, the surface area of the droplet population is calculated as the summation of the surface area of each individual droplet. The results of simulations with differing bin sizes are compared to the analytical equation in Figure B.4. The effect of gas phase saturation is clearly visible as the evaporation
Figure B.5: Non-dimensional surface areas plotted as a function of time for a bidisperse droplet population with (a) $d_{0,1} = 2 \ \mu m$, $N_{0,1} = 1 \times 10^{13}$ particles/kg and (b) $d_{0,2} = 10 \ \mu m$, $N_{0,2} = 1 \times 10^9$ particles/kg at $P_{v,0} = 0$, $T = 293 K$ and $P = 1$ bar.

The process eventually terminates at $t \approx 1.5$ ms.

Once again, it can be seen that as the bin size increases the deviation from the analytical equation increases. This is due to numerical diffusion. Using the analytical equation, all particles are the same size and decrease in size until the vapor has saturated the gas phase. In the numerical solution, particles move between bins in order to decrease in size and can only move to bins with pre-determined sizes. A portion of the particles move to a smaller bin each time step. This leads to some droplets completely evaporating, thereby lowering the total number of particles. The final populations for the analytical and numerical solution contain the same mass of liquid, however the numerical model has less particles which results in decreased surface area. This deviation can be seen in Figure B.4.

An analogous simulation is also run for a bidisperse water aerosol with $d_{0,1} = 2 \ \mu m$, $d_{0,2} = 10 \ \mu m$, $N_{0,1} = 1 \times 10^{13}$ particles/kg and $N_{0,2} = 1 \times 10^9$ particles/kg at $T = 300 K$ and $P = 1$ bar. Results are presented in Figure B.5 and indicate the same trends as the monodisperse aerosol. While the plots for the small and large droplets appear the same, the scale of the small droplet plot is much larger than the scale for the large droplet plot. For the $d_0 = 2 \ \mu m$ drops, the difference between the analytical solution and the $2 \ \mu m$ per bin simulation is 11 % while the difference is only 0.8 % for the larger droplets. This illustrates that that large droplets are less affected by numerical diffusion because they are farther away from the smallest bin and only lose a few droplets during the simulation.
Figure B.6: The relative humidity during the initial evaporation of \( n \)-dodecane in the mixing chamber using different values for the evaporation coefficient.

**Bin Size Selection**

Based on the results of these simulations a bin spacing of 0.1 \( \mu \)m is selected to be used in the aerosol model. Some deviation from the analytical solution is present, but decreasing the bin size further increases the computational cost of the model.

**Evaporation Coefficient**

The evaporation coefficient is used to determine the number of molecules that evaporate during a particular time step. In Eqs. (3.57) and (3.58) the value used in the aerosol model is determined by conducting simulations of \( n \)-dodecane in air at \( T = 350 \) K, \( P = 1 \) bar and \( \phi = 2 \). The coefficient is initially set at 0.2 and decreased until the vapor pressure does not over-shoot the saturation pressure. The relative humidity is monitored to determine when this occurs. The results are shown in Figure B.6.

Using a value of 0.2 for the evaporation coefficient results in the oversaturation of the vapor phase. This is because as liquid molecules evaporate, the temperature of the droplet decreases and the saturation pressure is lowered. Smaller steps need to be taken in order to ensure that the vapor pressure does not exceed the saturation pressure. As the evaporation coefficient is decreased, the amount of saturation pressure over shoot also decreases. Based on the results seen in Figure B.6 an evaporation coefficient of 0.02 is utilized in the aerosol model.
Figure B.7: Diameter under which 98% of the mass is present and aerosol losses as a function of number of discretizations for the (a) stirred vessel, (b) straight tube, (c) curved tube and (d) cyclone removal sub-models.

Coagulation Time Steps

The minimum number of time steps needed in each component of the delivery system is determined based on simulations using \( n \)-decane in air with \( Re = 200 \), \( \phi = 2 \), \( T = 300 \) K and \( P = 1 \) bar. These conditions are used because the aerosol will be retained the longest in the delivery system and will have the highest concentration of droplets. For higher flow rates, greater gas temperatures and lower fuel loadings (i.e. \( \phi \approx 0.5 \)) the modification to the DSD due to coagulation will be less. Results using the OnQ nebulizer distribution are discussed in this section.

The number of discretized sections in each component is initially set to one and the simulation of the delivery process is run. The actual number of divisions is set in the model. The number of divisions is then doubled and another simulation is run. This process is continued until there is either no significant change in the results, or the results are shown not to stabilize. The percent aerosol loss, and the droplet diameter under which 98% of the ‘fuel’ mass is contained, \( d_{98} \), are the metrics that are monitored.

The removal sub-models are first tested without coagulation or evaporation to
ensure that removal sub-models are stable. Components are tested individually using the OnQ nebulizer distribution for the initial incoming DSD. Plots from this series of simulations can be seen in Figure B.7. The removal sub-model for the mixing chamber provides stable results after about 10 time steps, or discretizations. The sub-models for aerosol loss in a straight tube and for the cyclone flow give results that are stable using only one time step. Finally, the aerosol loss for the tube bend removal sub-model shows that about 16 steps are needed before the value is stable.

After investigating the sub-models individually, the simulations are run with the coagulation and evaporation models in order to determine the minimum number of bins required to adequately resolve the aerosol dynamics. The components are tested in the
order that the aerosol moves through the system. Results from the simulations are shown in Figure B.8.

The maximum diameter and aerosol loss within the mixing chamber stabilize after about 60 time steps, or discretizations, while all of the other sub-models need 16 steps before the aerosol loss stabilizes. These values are used in the aerosol model as the minimum discretizations in each component.
Appendix C

TUBE BEND DEPOSITION

Due to discrepancies seen with many tube bend models (e.g. [48, 50, 53]) a correlation was developed using the Tsai and Pui [53] model where this was modified from the original form, and then validated with experimental gravitational measurements.

The results of simulations from Tsai and Pui [53] are fit using the Weibull distribution. This is necessary because the formulations provided in their paper are not sufficient for utilization over a wide range of conditions.

The Weibull distribution is selected based on the shape of the derivatives of the Tsai and Pui [53] deposition efficiency curves, which indicate similar behavior as the probability distribution functions (PDFs) of the Weibull distribution. The PDF of a Weibull distribution is

$$f(x) = \frac{c}{b} \left( \frac{x}{b} \right)^{c-1} e^{-\left(\frac{x}{b}\right)^c}$$  \hspace{1cm} \text{(C.1)}

when $x$, the independent variable, is greater than zero. The scale and shape parameters are $b$ and $c$, respectively. The cumulative distribution function (CDF) is the integral of the PDF and is used to represent the deposition efficiency curves from the paper

$$\chi_{b,LF} = 1 - e^{-\left(\frac{St}{b}\right)^c}$$  \hspace{1cm} \text{(C.2)}

where the Stokes number is the independent variable.

Six relevant curves are used from the paper and are manually fit so that the smaller Stokes numbers had the best fit. This is done to keep the relative error of the fit to a minimum. The droplets expected in the aerosol system have Stokes numbers below 0.1. The parameters for the curve fits are presented in Table C.1 and the CDFs are...
Table C.1: Parameters for Weibull distribution fits to deposition efficiency curves of Tsai and Pui [53].

<table>
<thead>
<tr>
<th>Re</th>
<th>De</th>
<th>Ro</th>
<th>b</th>
<th>c</th>
</tr>
</thead>
<tbody>
<tr>
<td>170</td>
<td>38</td>
<td>20</td>
<td>0.85</td>
<td>3.6</td>
</tr>
<tr>
<td>1000</td>
<td>419</td>
<td>5.7</td>
<td>0.38</td>
<td>2.8</td>
</tr>
<tr>
<td>1874</td>
<td>419</td>
<td>20</td>
<td>0.58</td>
<td>2.7</td>
</tr>
<tr>
<td>101</td>
<td>38</td>
<td>7</td>
<td>0.62</td>
<td>3.4</td>
</tr>
<tr>
<td>373</td>
<td>141</td>
<td>7</td>
<td>0.49</td>
<td>3.1</td>
</tr>
<tr>
<td>2999</td>
<td>869</td>
<td>7</td>
<td>0.38</td>
<td>2.8</td>
</tr>
</tbody>
</table>

Figure C.1: Weibull distribution fits (lines) to the deposition efficiency curves (points) from Tsai and Pui [53].
Figure C.2: Shape parameter correlations of (a) $f_1(De)$ and (b) $f_2(R_o)$.

compared to the deposition efficiency curves in Figure C.1.

The shape and scale parameters are then correlated to either the flow Reynolds number, Dean number and/or the curve ratio in order to interpolate the results. The shape parameter is first correlated to the Dean number using an exponential fit

$$f_1(De) = -0.1067\ln(De) + 1.1015$$  \hspace{1cm} (C.3)

Next, shape parameter is divided by $f_1(De)$ and correlated to the curve ratio using a linear fit

$$\frac{b}{f_1(De)} = f_2(R_o) = 0.0222R_o + 0.5944$$  \hspace{1cm} (C.4)

Using the two previous equations, the shape factor can be calculated by

$$b_{calc} = f_1(De)f_2(R_o)$$  \hspace{1cm} (C.5)

Plots illustrating the correlations are presented in Figure C.2.

A similar process is used to correlate the scale parameter. In this case, however, the correlations both utilize linear fits and are

$$g_1(R_o) = 0.0106R_o + 2.9493$$  \hspace{1cm} (C.6)

$$\frac{c}{g_1(R_o)} = g_2(Re) = -1.0772 \times 10^{-4}Re + 1.1056$$  \hspace{1cm} (C.7)
with the scale parameter calculated as

\[ c_{\text{calc}} = g_1(R_o) g_2(Re) \]  

Plots showing the correlations to the scale parameter are presented in Figure C.3.

The calculated and published [53] deposition efficiency curves are compared in Figure C.4. Overall, the curves compare well with a small overprediction when the Stokes number is above \( \approx 0.4 \). This is acceptable for this study because nearly all of the liquid mass is in droplets with Stokes numbers below this value.

Experiments are conducted to validate the new correlations where \( \approx 90\text{-degree} \) bend is utilized with \( R_o = 7.38 \). This radius is most applicable to the expected system configuration. The experiments were setup the same way as experiments conducted to measure the aerosol loss in the delivery system discussed in Chapter 4. In this case, the outlet of the cyclone mixing chamber is connected to a vertical piece of 0.43 m x 23.8 mm ID aluminum tube followed by a piece of 25.4 mm ID Tygon tubing which is fit into a jig made from a 2 x 12 piece of wood. The jig has a channel cut with turn radius of 94 mm and straight sections of 25.7 mm before and after the turn. The tube is pressed into the jig using a C-clamp to prevent the tube from pinching. The aerosol (air + water) enters the curve vertically flowing with gravity and exits the bend horizontally. This can be seen in Figure C.5.

A range of Dean numbers is investigated during these validation tests with the
Figure C.4: A comparison of deposition efficiency curves calculated (lines) using Eqs. (C.2), (C.5) and (C.8) with results (points) from Tsai and Pui [53].

Figure C.5: Side and cross-section views of the deposition observed in a 90 degree tube bend at $R_o = 7.38$ and $Dn = 484$. The dashed line indicates the boundary where deposition occurs.
Figure C.6: Comparison between measured and modeled aerosol losses in a tube bend at $T = 297$ K, $P = 1$ bar and low flow OnQ DSD. The model only accounts for losses due to the flow and significantly underpredicts losses.

measurements shown in Figure C.6 along with predicted deposition loss using the new correlation. The DSD for the low flow OnQ nebulizer is used in the calculations. The trend in the experimental results indicates that as the gas flow rate is reduced and the Dean number decreases the aerosol loss in the bend increases. This, however, is opposite of the trend seen in the model results. The figure also indicates that the correlated model significantly underpredicts the aerosol loss in the bend. This could be due to the fact that the model developed by Tsai and Pui [53] and others (e.g. [48, 50]) only account for losses due to the flow field.

The deposition pattern observed in the tube bend highlighted in Figure C.5 suggests that gravity also significantly influences the aerosol loss and indicates that this must be taken into account when modeling the bend loss. The importance of this effect was also demonstrated by Gutfinger et al. [95]. When viewing the tube from the side after conducting the experiment, as shown in Figure C.5, a distinct zone of deposition and one of no deposition are visible. When viewing the tube cross-section, the deposition of the water distinctly stops where the perimeter of the tube begins to curve towards the top. If the deposition were due to the flow alone, it is expected that no distinct deposition zone would form because the different sizes of aerosol particles would have different deposition patterns. The overlapping of these patterns should result in gradual transitions between
Figure C.7: Comparison between measured and modeled aerosol losses in a tube bend at $T = 297$ K, $P = 1$ bar and low flow OnQ DSD. The model accounts for losses due to the flow and gravity.

The deposition and deposition free portions of the tube wall. It is also expected that some droplets would deposit on the inside edge (i.e. the top of the cross-section) of the bend, however this is not observed during the experiments.

To account for gravity, the sub-model for straight tube deposition presented in Section 3.5 is combined with the modified sub-model of Tsai and Pui [53]. The angle of the tube is based on the line tangent to each control volume of the tube bend. Results from the combined sub-models are shown in Figure C.7. The new model still underpredicts the experiment results, but the values are closer and follow the same trend.

One reason that the model predictions do not match the experiment results is that the droplet distribution of the low flow OnQ nebulizer may not be correct. In order to test this hypothesis, simulations were run with the combined model using the high flow OnQ nebulizer distribution and the results are shown in Figure C.8. Once again, the loss in the bend is still underpredicted.

The difference between the models and experiment results could be due to effects of gravity on the flow field and the coagulation as larger drops move outward toward the walls. Both of these mechanisms would require extensive work in order to develop and validate an appropriate model. In an attempt to match the experiment results, the models for gravity (i.e. Anand et al. [8, 43]) and flow (i.e. Tsai and Pui [53]) were multiplied by a
Figure C.8: Comparison between measured and modeled aerosol losses in a tube bend at $T = 297 \, \text{K}$, $P = 1 \, \text{bar}$ and high flow OnQ DSD. The model accounts for losses due to the flow and gravity.

Figure C.9: Comparison between measured and modeled aerosol losses in a tube bend at $T = 297 \, \text{K}$, $P = 1 \, \text{bar}$ and low flow OnQ DSD. The model accounts for losses due to the flow and gravity and sub-models were adjusted by the factor listed in the legend.
factor of 3.75 and 10 respectively. The factors were applied one at a time and were determined by matching the results from the experiment with a Dean number of 876. Results from the adjusted models are shown in Figure C.9. It can be seen that when the sub-model that accounts for gravity is multiplied by a factor of 3.5, the experiment results are predicted fairly well. Therefore, the sub-model for bend removal in the aerosol model is

$$\chi_b = \frac{\theta_b}{90} \chi_{b,LF} + 3.75 \chi_{tu}$$  \hspace{1cm} (C.9)

The sub-model assumes uniform deposition throughout the bend. It was developed over a range of Dean numbers with $R_o = 7.38$ and the bend is positioned such that the inlet is above a horizontal outlet. The bend angle, $\theta_b$ is measured in degrees.
Appendix D

UNCERTAINTY

The uncertainty associated with experimental gravimetric measurements is determined using the data in Table D.1 [83]. Measurements are conducted using the system with the 0.152 m diameter cyclone discussed in Section 4.3 at 297 K and 1 bar with 3 nebulizers and inlet manifold #5. Losses from the mixing chamber and delivery tube were analyzed because these components were the same for each test. It is assumed that the error associated with losses in the inlet manifold and reaction chamber are of similar magnitude. The losses in these components were not analyzed because the valve position in the inlet manifold was different in each test. Standard statistical analysis produced an absolute error of 0.48 % at a 95 % confidence level for the cyclone and delivery tube loss measurements.

The error associated with the aerosol losses is also calculated by propagating the measurement uncertainty of the instruments. The resolution of the scale, pressure gage, timer and thermometer are 0.1 g, 0.1 psi, 1 s and 0.1 C, respectively. Using EES, the absolute error associated with aerosol loss measurements is calculated to be 0.36 % in each component. This value matches well with the statistical analysis of the repeated

Table D.1: Measurements of aerosol losses in the mixing chamber and delivery tube for similar systems.

<table>
<thead>
<tr>
<th>Test</th>
<th>1</th>
<th>2</th>
<th>3</th>
<th>4</th>
<th>5</th>
<th>6</th>
<th>7</th>
</tr>
</thead>
<tbody>
<tr>
<td>Air Flow Rate (L/min)</td>
<td>19.8</td>
<td>17.3</td>
<td>16.6</td>
<td>16.6</td>
<td>17.9</td>
<td>20.0</td>
<td>20.0</td>
</tr>
<tr>
<td>Water Flow Rate (g/min)</td>
<td>0.94</td>
<td>0.95</td>
<td>0.97</td>
<td>0.97</td>
<td>0.97</td>
<td>0.98</td>
<td>1.09</td>
</tr>
<tr>
<td>Loss in Cyclone (%)</td>
<td>2.12</td>
<td>2.46</td>
<td>2.05</td>
<td>2.75</td>
<td>1.72</td>
<td>3.07</td>
<td>3.07</td>
</tr>
<tr>
<td>Loss in Delivery Tube (%)</td>
<td>2.12</td>
<td>2.46</td>
<td>2.74</td>
<td>3.09</td>
<td>2.06</td>
<td>3.41</td>
<td>2.15</td>
</tr>
</tbody>
</table>
experiments and shows that 75% of the random error is associated with the measurement equipment.

Uncertainties for the individual components are summed to calculate the uncertainty of total measured aerosol loss in a system. Therefore, the absolute uncertainty of the total loss is equal to 1.92%.
Appendix E

SHADOWGRAPHY AND PDA RESULTS

Showdowgraphy and phase doppler anemometry (PDA) are two techniques for measuring fine DSDs of liquid droplets suspended is a gas phase or gas bubbles suspended in a liquid phase. The intent of the measurements performed for this thesis was to provide and additional means of validation for the numerical model.

The setup to take shadowgraphy measurements is shown in Figure E.1. The droplets pass between a backlight source and an image capturing device. The image captured is the shadow that is created by the droplets as they pass in front of the light source. An algorithm is then used to determine the size of the droplets based on the image collected.

Measurements for the current study were carried out using the SizingMaster Shadow package from LaVision. The equipment used was:

- Navitar 12 x zoom

Figure E.1: Diagram of a Shadowgraphy measurement setup [96].
Figure E.2: Droplet measurements from a) 0 - 20 μm and b) 5 - 20 μm comparing shadowgraphy and PDA measurements against the results of Kuhli et al. [85].

- Mitutoyo 5 x zoom lens
- LaVision Diffuser
- Calibration disk
- Imager pro X CCD camera
- New Wave Research Solo PIV model: Solo II-15 Hz
- Ekspla NL 303D/SH/TH/FH
- Ekspla PS 5012E Power Supply
- Berkeley Nucleonics Corporation Model 565 Pulse/Delay Generator

Two laser sources were used, SoloPIV and Ekspla, with no difference in results. Images were collected using DaVis 7.2 software and processed using the SizingMaster Shadow software package. For a full description of measurement setup and image processing see the product manual for the SizingMaster Shadow software package.

Measurements were taken of the droplets created by an Aeroneb OnQ nebulizer and compared with the results from Kuhli et al. [85]. For shadowgraphy measurements, many variables were adjusted during image processing to try to improve the measurement of droplets below 5 μm. The best results can be seen in Figure E.2. Figure E.2a shows the distribution of droplets from 0 - 20 μm while Figure E.2b shows only the droplets between 5 - 20 μm. As can be seen here, the droplets smaller than 5 μm were not measured accurately by the shadowgraphy system. Upon further review of the product manual, it was found that the shadowgraphy technique is intended for droplets larger than 5 μm.
A typical PDA setup is shown in Figure E.3. A laser is split into two beams which intersect to form an interference pattern. As a droplet travels through the pattern, the scattered light is collected by photodetectors and the size of the droplet is computed [97]. Probe volumes are very small and measure one droplet at a time as opposed to shadowgraphy, where images capture a larger area containing multiple droplets.

PDA measurements were taken at the Multiphase Flow Research Lab at the University of Illinois Urbana-Champaign using the OnQ low VMD nebulizer. As shown in Figure E.2, measurements did not match the published data, and due to limited access to the equipment, there was not an opportunity to optimize the measurement parameters. It has been shown that the operating conditions of the equipment (e.g. beam power, photomultiplier voltage, etc.) must be carefully chosen in order to accurately measure small droplets [98], and this may be a reason for the discrepancy between the published and measured DSDs.

In order to investigate the range of particles of interest in this study (0.1 - 20 um), an optimized PDA system, or another method such as laser diffraction would need to be used.