COMPUTATIONAL INVESTIGATIONS OF POLYMER SHEET BREAKUP
FOR OPTIMIZATION OF DEVOLATILIZATION PROCESSES IN STEAM
CONTACTORS

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COMPUTATIONAL INVESTIGATIONS OF POLYMER SHEET BREAKUP
FOR OPTIMIZATION OF DEVOLATILIZATION PROCESSES IN STEAM
CONTACTORS

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Thesis

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ABSTRACT

Polymer devolatilization is a vital process in polymer manufacturing and is significantly impactful on the successful creation of high quality polymers, meeting both rigorous product specifications and regulatory requirements. Polymers resulting from such processes have wide applications ranging from agricultural and biomedical solutions to aerospace components and even to modern day clothing and accessories.

Although there are several popular methods used to accomplish the devolatilization process, this research focuses specifically on steam stripping, where superheated steam is used to remove any unwanted substances, such as volatiles and solvents, from the polymer mixture. This polymer mixture, referred to as “cement” and comprised of polymer and a cyclohexane solvent, undergoes mixing with superheated steam in a contactor to evaporate and remove the cyclohexane. Between the heat and the aerodynamic forces caused by the mixing, the liquid polymer experiences sheet breakup.

The objective of the current study is to create a computational fluid dynamics (CFD) model that solves for the initial breakup of the liquid mixture, and then use the resulting diameter distribution to simulate the trajectory and multiphase mass transfer of the cement as it forms into smaller and smaller droplets. A parametric
study is conducted in order to determine the effects of contactor geometry changes
on the initial sheet breakup and the resulting impacts to the final polymer product
quality. The purpose for modifying the geometry is to increase the uniformity of the
breakup as well as reduce the amount of cement that sticks to the contactor walls.
The use of CFD allows the industry partner for this research to try multiple different
optimization solutions without interrupting production and spending large amounts
of money on trial-and-error prototypes.
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Finally, I cannot express enough love and gratitude to my wife and children who have faithfully supported and encouraged my dream of someday also educating future generations of engineers.
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1.1 Problem Overview

Polymers pervade almost every facet of modern day life, being found in industry products from automobiles and airplanes to biomedical devices to packaging to even waterproof coatings for clothing. Even the American comedian, Jon Stewart, used polymer composition as a well-understood metaphor to describe his prior satirical television show saying, "That’s the beauty of our show. Comedy or politics. We’re sort of a mix. A space age polymer of both. A synthetic comedy-like material." [1]

The quality of each polymer is paramount to the successful use of the material for its intended application. High quality polymers, in particular, require significant refinement to meet product specifications and environmental regulations. Perhaps the most significant manufacturing process affecting quality is the process of polymer devolatilization. Essentially, polymer devolatilization is the removal of unwanted byproducts in the polymer mixture, such as monomers, solvents, and any other volatile products introduced when making the polymer [2].

There are variety of methods used in industry to accomplish this process, but this research focuses on steam stripping, where superheated steam is mixed with
the polymer mixture to evaporate unwanted solvents. The devolatilization process occurs inside the steam contactor, a pressurized vessel operated under the necessary pressures, temperatures, and velocities to cause the polymer mixture to disintegrate into smaller liquid structures or droplets. These droplets continue to break apart as they are carried through the contactor by the superheated steam, which also serves as the heat source necessary to further evaporate the solvents. The resulting product mixture, comprised of a higher polymer content and a lower volatile content than at the start of the process, then undergoes additional processing to create granular polymer particles called "crumb."

This thesis evaluates a very specific application of this process based on contactor design and operating conditions established by an industry partner and world leading manufacturer of several different types of polymers. Previous research by the same authors has focused on creating a computational fluid dynamic (CFD) model to simulate flow conditions as they are found in the manufacturing facility [3, 4]. The model of this multiphysics process was used to optimize the consumption of costly superheated steam and evaluate the affects of geometry changes on droplet evaporation in the mixing and nanogap zones of the contactor [5, 6]. The intent of this thesis is to further build on that research by assessing the impact of contactor design on the initial breakup of the polymer mixture near its inlet and on how much polymer mixture sticks to the walls of the vessel.

The liquid polymer mixture in this research, hereon referred to as "cement," is comprised of polymer and cyclohexane, a hydrocarbon solvent. The superheated
steam and cement are both injected into the annular space of the contactor via circumferential slits. The cement is injected downstream of the steam. The resulting interaction can be categorized into two different multiphase flow phenomenon: primary and secondary sheet breakup. The first occurs in the section of the contactor nearest the inlets as the cement comes into contact with the high pressure and temperature steam. Upon contact, the liquid column begins to shear apart into liquid ligaments and particles. The second phenomena occurs downstream as the droplets continue to experience heat transfer and aerodynamic forces from the steam, leading to further potential breakup and evaporation of the cyclohexane.

Because trial-and-error changes to the actual contactor, known as the G1 steam contactor, and operating conditions at the manufacturing facility would result in higher costs and loss of productivity, CFD is a very cost-effective tool to evaluate a number of different modifications. The complexity and computational expense associated with modeling the multiphase flow interaction necessitates the development of a separate model for the secondary, multiphase flow regime. The model, created using the commercial CFD software ANSYS Fluent, is tuned to match actual production data from the industry partner [5]. Then geometry and operating conditions are modified to optimize the final product quality, specifically looking at and outlet particle properties and volume of cement exiting the contactor against the walls.
1.2 Scope

The intent of this research, as documented in this thesis, is to accurately model the polymer devolatilization process and the effects of contactor design changes on the final polymer product quality. Because of the complexity of the devolatilization process, the production facility could implement many different contactor design changes without seeing positive effects. Therefore, the use of CFD modeling is particularly effective at providing a high confidence, lower cost solution.

In this study, the effect of the geometry modifications to the contactor are evaluated using two different CFD models: the first is used to measure the effect on primary sheet breakup and initial droplet diameter distribution, and the second tracks secondary phase change of those same particles through the contactor, neglecting for now the potential for further breakup downstream [5]. Each simulation is intended to optimize the primary sheet breakup and reduce the amount of polymer mixture sticking to the contactor walls, so that the final polymer crumb consistently meets end-state cyclohexane content specifications.

1.3 Literature Review

While there is little research available that corresponds directly to the topic of polymer devolatilization and sheet breakup, the numerical modeling of sheet breakup is a common study in other relevant applications. The study of polymers began early in the 19th century with the first synthetic polymer being developed in 1907 [7]. The
The most significant contribution to polymer science was provided by Hermann Staudinger who won the Nobel Prize in 1953 for being the first to suggest that polymers were actually atoms held together by covalent bonds [8].

Over the next 30 years, the industrial growth of polymer manufacturing led to a prolific establishment of academic programs and research centers around the world, including one of the earlier pioneering programs at The University of Akron’s College of Polymer Science and Polymer Engineering in 1988.

1.3.1 Polymer Devolatilization

While most academic research has focused on the development of improved or new polymer properties, industry has addressed the challenges associated with manufacturing, namely the presence of undesirable volatiles. Most polymers contain low-molecular weight components like unreacted monomers, solvents, and other by-products. These are collectively known as volatiles and are separated from the polymer for a variety of reasons, including regulatory requirements, polymer quality, and odor elimination [2].

This separation process is known as polymer devolatilization and can be accomplished utilizing either static or dynamic equipment, depending on the polymer mixture composition (e.g. volatile mass fraction) [9, 10]. Static methods, as in this research, are particularly effective for the devolatilization of low-viscosity, high volatile mass fraction mixtures by causing the solvent to evaporate rapidly or flash [2, 5]. The flash occurs when the vapor pressure, \( p^* \), of the volatile in the polymer
mixture exceeds its own local partial pressure, \( p_v \) [11]. Because the vapor pressure is heavily dependent on temperature, the operating temperature of the polymer mixture must be high enough (without resulting in thermal degradation) to allow \( p^* - p_v > 0 \) upon injection into the static devolatilization equipment. The temperature increase is provided by a heated stripping agent like the superheated steam in this research.

Despite being used for its simplicity and lower operating costs, the geometry and operating conditions of the static equipment must be optimized to prevent over- or under-exposure to the heated stripping agent and inconsistent final product quality [5].

1.3.2 Primary Liquid Sheet Breakup

When the cement enters the steam contactor to begin the devolatilization process, it begins to break apart as a result of both the evaporation of solvent and the shear created by the steam flow. This breakup phenomenon is also referred to as liquid atomization and has been the subject of prevalent research over the last several decades. Very few of these studies in literature are specifically related to the polymer devolatilization process. Instead, much of the available research on sheet breakup is in support of other relevant industrial and manufacturing applications, like combustion engine fuel injection, metal powder formation, agricultural sprays, and rocket engines.

The liquid atomization process occurs in two consecutive phases known as primary and secondary breakup. Primary breakup usually begins at the jet surface
where the gas impacts or contacts the liquid stream, causing it to disintegrate into ligaments, droplets, and other liquid structures of varying size [12, 13]. The larger liquid structures may further break up into smaller and smaller droplets during the secondary breakup phase.

Primary breakup typically occurs in two distinct, consecutive stages. The first stage is brought on by an initial disturbance at the jet surface, which is further increased at the second stage by the aerodynamic forces created by the high momentum of the gas flow [14, 15]. Substantial research has been devoted to understanding the cause of the initial perturbation for both laminar and turbulent flows and has mainly focused on the analysis of the instabilities occurring at the exit of the injection point and the associated wavelengths. For laminar liquid flow, Kelvin-Helmholtz instability creates the initial disturbance, connecting the instability wavelength magnitude and the boundary layer formed between the flowing gas and liquid nozzle. In the second stage of primary breakup, the disturbances are further magnified by Rayleigh-Taylor instabilities caused by the drag and other aerodynamics forces from the gas acting on the injected liquid [16]. In the case of a turbulent jet discharge, the incited instabilities are the direct result of eddies formed within the liquid [17].

Research addresses two common geometric configurations for primary breakup: coaxial and cross gaseous flow. Both types have been heavily studies to understand the the sensitivities to changes in fluid properties [18, 19]. While liquid properties are generally considered to have little effect on the nature of breakup regimes themselves, available research does show that they have a significant impact on the actual
breakup length and time - how far and how long the liquid jet travels before beginning to break up [20]. The most common material properties considered are viscosity and surface tension. The correlation between these properties is used to characterize the breakup via several non-dimensional parameters.

For coaxial flow, visually shown in Figure 1.1, the gas Weber number, liquid Reynolds number, momentum flux ratio, and momentum ratio are used to describe the atomization process:

\[
W_G = \frac{\rho_G(U_G - U_L)^2D_L}{\sigma}
\]

\[
Re_L = \frac{\rho_L U_L D_L}{\mu_L}
\]

where \( \rho_L, U_L, \) and \( \mu_L \) are the density, velocity, and viscosity of the liquid; \( \rho_G \) and \( U_G \) density and velocity of the gas; and \( D_L \) and \( \sigma \) are the liquid jet diameter and surface tension coefficient, respectively [21]. Increasing the liquid Reynolds number
Figure 1.2: Schematic of a turbulent liquid jet in crossflow illustrating a typical breakup scenario

and Weber number leads to increasingly sudden and fine disintegration of the liquid structures immediately following the jet exit.

For cross flow applications, shown in Figure 1.2, are the primary focus of this research [12]. In these instances, the gaseous Weber number is again considered along with the liquid/gas momentum flux [12, 22]:

\[ We_G = \frac{\rho_G U_G^2 D_L}{\sigma} \]  \hspace{1cm} (1.3)

\[ q = \frac{\rho_L V_L^2}{\rho_G U_G^2} \]  \hspace{1cm} (1.4)

At lower liquid jet Weber numbers and momentum fluxes, the liquid exhibits a column-style breakup, maintaining its original connectivity further from the jet exit. The liquid structures break off from the end of the liquid stream. However, as the jet Weber number and momentum flux increase, the breakup transitions to a bag-type breakup, which appears as a fine spray and occurs very near the jet exit. Both types of transitions will be exhibited in this thesis.
1.3.3 Coupled Level-Set Volume of Fluid

The modeling of primary breakup has historically been quite difficult, because the simulation is required to account for multiphase flow under significant aerodynamic forces and turbulent effects \([14, 23]\). Up until the last few decades, most research has focused on relatively small liquid/gas density ratios of less than 100. The accuracy of those numerical models begins to drift for high density ratios \(O(1000)\) due to the discontinuities of density and viscosity, causing divergence in the pressure equation convergence \([24]\). It has only been within the last 5-7 years that viable modeling solutions have begun to close the gap between experimental and simulation results for these conditions. While there has been a variety of unique models proposed to solve this complex problem, the most successful simulations define the interface implicitly through a separate variable whose solution comes from the Eulerian transport equation \([13]\). The two most widely researched are the Volume of Fluid (VOF) and Level Set (LS) methods.

Developed more than 30 years ago, the VOF model has been successfully used in numerous applications of primary breakup and is particularly effective at mass conservation \([25, 24]\). Defined as the volume fraction of liquid in each computational cell, the VOF function \(F\) ranges between 0 and 1. The volume fraction is described by:

\[
\frac{\partial F}{\partial t} + U \cdot \nabla F = 0
\]  

(1.5)
where

\[
F = \begin{cases}
1 & \text{if cell is fully occupied by liquid} \\
0 & \text{if cell is fully occupied by gas}
\end{cases}
\quad (1.6)
\]

and if the cell has both gas and liquid, then \(0 < F < 1\). Unfortunately, because the liquid fraction is defined for each cell, the quality of the interface geometry (e.g. curvature \([26]\)) is dependent on an increasingly fine but computationally expensive grid, particularly for 3D models.

To improve interface tracking, the LS method was created. Whereas the VOF function \(F\) is discontinuous, the LS function, \(\phi\), is a continuous variable, capturing the interface geometry much more effectively than the VOF method. The function is defined by:

\[
\frac{\partial \phi}{\partial t} + U \cdot \nabla \phi = 0
\quad (1.7)
\]

where

\[
\phi = \begin{cases}
0 & \text{if phase interface} \\
> 0 & \text{if liquid} \\
< 0 & \text{if gas}
\end{cases}
\quad (1.8)
\]

The biggest downside to the LS method is its weakness at mass conservation that, which again is largely resolved by a sufficiently fine mesh. While multiple refinements to the LS method have been created by shaping the LS function and through grid refinement techniques, the mass loss/gain can still be significant \([27]\).

In an effort to combine the best qualities of each method, the Coupled Level Set Volume of Fluid (CLSVOF) method was proposed at the turn of the century
Table 1.1: Mass error comparison

<table>
<thead>
<tr>
<th>Methods</th>
<th>LS</th>
<th>VOF</th>
<th>CLSVOF</th>
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<tbody>
<tr>
<td>Mass error</td>
<td>31%</td>
<td>$6 \times 10^{-4} %$</td>
<td>$5 \times 10^{-4} %$</td>
</tr>
</tbody>
</table>

[28]. This was further demonstrated by Xiao in 2012 who captured an informative side-by-side comparison of the three methods in Table 1.1 [13]. The success of this method is achieved by combining the interface normal vector from the LS method and the position in the cell from the VOF method. This optimum pairing is further detailed in the flow chart for the CLSVOF method shown in Figure 1.3 [29].

Figure 1.3: Illustration of the level set function
Numerous studies, like what is visually depicted in Figure 1.4, have been conducted to compare each of these methods to prove both the accuracy and simulation ease of this method [13, 30]. It is widely accepted from results such as these that the CSLVOF is most effective because it is (1) computationally efficient for 3D models, (2) proficient in resolving thin liquid structures, and (3) highly accurate in mass conservation [13].

1.3.4 Multiphase Droplet Flow With Lagrangian Tracking Approach

The secondary phase change simulation tracks the cement particle locations and phase change between the cement and superheated steam. The concentration of the particles in the contactor, therefore, has a significant affect on the interaction between the two phases. Depending on the concentration level, the flow can be tracked using either an Eulerian-Eulerian (E-E) or Eulerian-Lagrangian (E-L) frame of reference [31, 32, 33]. The E-L approach treats the fluid phase as a continuum and tracks the dispersed phase individually in a discrete phase. This makes it a highly accurate method for
tracking particles, especially when there is physical or thermodynamic interaction between them [34].

Unfortunately, as the particle density increases, the Lagrangian approach becomes increasingly computationally expensive compared to the E-E approach, which treats both phases as part of a continuum [35]. The E-L approach is most effective when the dispersed phase has a volume fraction less than 5% such that particle-particle interactions can be neglected [36, 37]. Since the secondary phase in this thesis is relatively dilute (approximately 3%), the E-L approach is a suitable choice [5]. Because of the requirements of the E-L approach, a Discrete Phase Model (DPM) is used to simulate the flow of the particles, modeling the continuous phase (steam) in an Eulerian frame and the discrete phase (polymer and solvent particles) in a Lagrangian frame. Each individual particle is tracked throughout the flow field.
CHAPTER II

PROBLEM DESCRIPTION

As mentioned in the scope of the thesis, this research is centered around a steam contactor, known as the G1 contactor, currently in production use at a polymer manufacturing facility. Previous research on the G1 contactor proposed geometry changes in the downstream area of the contactor in order to optimize secondary breakup and reduce steam consumption [3, 4, 5]. This study evaluates several geometric configurations of the steam and cement inlets in an attempt to enhance primary breakup and reduce the amount of cement that adheres to the contactor walls.

2.1 Original G1 Contactor

The G1 steam contactor is a pressurized vessel where superheated steam and cement are mixed to accomplish the polymer devolatilization process. The dimensions of the G1 steam contactor were provided by the industry partner. Figure 2.1 shows a cross-sectional view of the contactor, where the annular space is created by a plug (shown as hollow space in the figure) that fills the center of the contactor [5]. The total length of the contactor is 1320 millimeters with a maximum external diameter of 183 millimeters.
The process begins with the injection of superheated steam through a circumferential, annular slit at a 45° angle. Just downstream of the steam injection, cement is also injected into the annular space of the contactor through a circumferential slit. Injected normal to the contactor wall and into the oncoming flow of steam, the cement is immediately affected by the heat and aerodynamic forces of the steam. Because the composition is 86% cyclohexane solvent and 14% polymer, the solvent begins to flash upon contact, starting the disintegration of the cement liquid column.

To balance computational efficiency and accuracy, primary breakup and the resulting multiphase flow after breakup are evaluated in two different models. Primary breakup is evaluated using a full three-dimensional model of the first 254 millimeters of the contactor shown in Figure 2.3. A cutaway of this section is also displayed to show the annular space where the fluids are being injected. Secondary breakup is measured via an axisymmetric three-dimensional model of the entire contactor as shown in Figure 2.2 [5].
Figure 2.2: Geometry callouts for (Left) steam and cement inlets, (Center) symmetry boundary, and (Right) outlet.

Figure 2.3: Full 3D 254 mm section of the contactor used to solve for primary sheet breakup and a plane schematic view of the bottom near the inlets.
Table 2.1: Operating and boundary conditions of original geometry for G1 contactor

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Steam Inlet</th>
<th>Cement Inlet</th>
<th>Outlet</th>
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<tbody>
<tr>
<td>Velocity</td>
<td>—</td>
<td>3.16 m/s</td>
<td>—</td>
</tr>
<tr>
<td>Pressure (CSLVOF)</td>
<td><strong>1410 kPa</strong></td>
<td>—</td>
<td>1380 kPa</td>
</tr>
<tr>
<td>Pressure (DPM)</td>
<td>2100 kPa</td>
<td>—</td>
<td><strong>140 kPa</strong></td>
</tr>
<tr>
<td>Temperature</td>
<td><strong>599 K</strong></td>
<td>433 K</td>
<td>599 K</td>
</tr>
<tr>
<td>Temperature (DPM)</td>
<td><strong>599 K</strong></td>
<td>433 K</td>
<td><strong>366 K</strong></td>
</tr>
<tr>
<td>Boundary</td>
<td>Pressure Inlet</td>
<td>Velocity Inlet</td>
<td>Pressure Outlet</td>
</tr>
</tbody>
</table>

The industry partner for this research provided all operating conditions (in bold) in Table 2.1 for the original G1 contactor based on current production settings. The boundary conditions were chosen based on prior research and results obtained through this study, which are described later in the thesis [5].

2.2 Geometric Variations

After the model for the original geometry is created and validated with actual production measurements, the optimization opportunities are visually apparent in the simulation, namely inconsistent primary breakup and excessive cement sticking to the contactor walls. The objective then becomes to increase heat transfer between the steam and cement and to modify aerodynamics forces of the steam to enhance breakup while keeping the flow in the middle of the annular space. To achieve this,
three different geometries with correlating operating and boundary conditions are evaluated. In each case, boundary conditions are modified in order to maintain necessary production parameters, namely a cement mass flow rate of 3.9 \( kg/s \) and a steam-to-cement ratio between 0.6 and 1.0. Lower ratios are optimal due to the cost savings associated with reducing the use of superheated steam [5]. Once the flow becomes relatively uniform, several snapshots of the cyclohexane volume fraction images are taken at arbitrary points in time in order to calculate a particle diameter distribution as an input for the simulations measuring the resulting phase change further downstream. The results of these simulations for each geometry are used to measure the final polymer quality at the outlet.

2.2.1 Dual Steam Inlets

The first geometric modification to the original contactor design includes the addition of another steam inlet just downstream of the cement inlet. Depicted by Figure 2.4, this inlet is intended to add additional heat to the flow and prevent the upstream steam inlet from pressing the cement onto the wall. Like the upstream steam inlet, the downstream inlet is a circumferential, annular slit with the same operating and boundary conditions.

This additional inlet is modeled in two distinct ways: (1) steam injected normal to the contactor walls and (2) steam injected at a 45° angle like the upstream steam inlet.
2.2.2 Reversed Inlets

In an effort to inject the cement into the middle of conditioned steam flow, the second geometric modification shown in Figure 2.5 reverses the order of the cement and steam inlets, injecting the cement normal to the contactor walls. To force the steam to the middle of the contactor and take advantage of additional steam contact, another steam inlet is added on the other side of the annular space. Each steam inlet injects the steam at a 45° angle in the direction of the contactor flow path. All of the steam inlets share the same operating and boundary conditions as in the original geometry.

Figure 2.4: Cross-sectional view of 254 mm section of the contactor with dual steam inlets and plane schematic views of the bottom near the inlets
Figure 2.5: Cross-sectional view of 254 mm section of the contactor with reversed order of cement and dual, opposing steam inlets and a plane schematic view of the bottom near the inlets
Chapter III

Formulation and Governing Equations

3.1 Conservation Equations

When modeling fluid flow, the dimensionless parameter Reynolds number, $Re$, is used to characterize the flow. $Re$ is defined as:

$$Re = \frac{\rho VL}{\mu} \quad (3.1)$$

where $\rho$ is the density, $V$ is the fluid velocity, $L$ is the characteristic length, and $\mu$ is the kinematic viscosity of the fluid. The $Re$ calculated in this study is 14,008, and hence the flow is considered to be turbulent. Turbulent flows are characterized by fluctuating velocity fields, which further cause other transported quantities to fluctuate as well at high frequency. So the instantaneous conservation equations can be time-averaged to remove the small scales, resulting in a modified set of equations that are computationally less expensive to solve [38, 39]. In addition, due to the high steam velocity near the converging-diverging gap in the steam contactor, the flow is compressible, with the Mach number, $Ma = V/c = 2.1$, being supersonic after the gap. In order to account for the density and temperature fluctuations, the compressible Navier-Stokes equations, in this case, undergo a density-weighted
time-averaging or a Favre-averaging procedure given by:

\[
\langle \ldots \rangle = \frac{\overline{\ldots}}{\rho}
\]  

(3.2)

where the overbear denotes the conventional Reynolds-average. So the Favre-averaged Navier-Stokes (FANS) conservation equations are given by:

\[
\frac{\partial \rho}{\partial t} + \frac{\partial}{\partial x_i} (\rho \overline{u_i}) = 0
\]  

(3.3)

\[
\frac{\partial (\rho \overline{u_i})}{\partial t} + \frac{\partial}{\partial x_i} (\rho \overline{u_j} \overline{u_i}) = -\frac{\partial P}{\partial x_i} + \frac{\partial}{\partial x_j} \left[ \overline{t_{ji}} - \rho \overline{u_i^\prime} \overline{u_i^\prime} \right]
\]  

(3.4)

\[
\frac{\partial}{\partial t} \left[ \rho \left( e + \frac{\overline{u_i} \overline{u_i}}{2} \right) + \frac{\rho \overline{u_i^\prime} \overline{u_i^\prime}}{2} \right] + \frac{\partial}{\partial x_j} \left[ \rho \overline{u_j} \left( h + \frac{\overline{u_i} \overline{u_i}}{2} \right) + \overline{u_j} \rho \overline{u_i^\prime} \overline{u_i^\prime} \right]
\]

\[
= \frac{\partial}{\partial x_j} \left[ -q_{Lj} - \rho \overline{u_j^\prime} h^\prime + t_{ji} \overline{u_i^\prime} - \rho \overline{u_j^\prime} \overline{u_i^\prime} \overline{u_i^\prime} \right] + \frac{\partial}{\partial x_j} \left[ \overline{u_i} \left( t_{ij} - \rho \overline{u_i^\prime} \overline{u_i^\prime} \right) \right]
\]  

(3.5)

\[
P = \rho R \tilde{T}
\]  

(3.6)

In Equations 3.3, 3.4, and 3.5, \((\ldots)^\prime\) represents the fluctuating quantity [40], [41]. \(\rho\) is the density, \(u\) is the velocity, \(P\) is the pressure, \(t_{ji}\) is the viscous stress tensor, \(e\) and \(h\) are the specific internal energy and specific enthalpy, respectively, and \(q_{Lj}\) is the laminar mean heat flux. Equation 3.6 is the Favre-averaged ideal gas equation used to model the continuous steam phase. In this problem, the \(Ma\) attained inside of the contactor is above 1, which makes the flow compressible, and hence modeling the steam as an ideal gas is apropos. The viscous stress tensor is written as:

\[
t_{ji} = 2\mu \delta_{ij} + \frac{\partial u_k}{\partial x_k} \delta_{ij}
\]  

(3.7)
where \( s_{ij} \) is the strain rate tensor, \( \zeta \) is the second viscosity coefficient, and \( \delta_{ij} \) is the Kronecker delta. The strain rate tensor is written as:

\[
s_{ij} = \frac{1}{2} \left( \frac{\partial u_i}{\partial x_j} + \frac{\partial u_j}{\partial x_i} \right).
\]

(3.8)

3.2 Turbulence

With regard to the turbulence model, the RNG \( k - \epsilon \) model is employed in the simulations presented here. The RNG \( k - \epsilon \) model is employed due to its improved accuracy for capturing the effect of swirl on turbulence, and flows with strong streamline curvatures, vorticies, and rotation. This simulation shows regions of highly strained flow near the gap region, thus the choice of this model is appropriate for this simulation [42, 43, 44, 45]. The compressible RNG \( k - \epsilon \) transport equations are written as:

\[
\begin{align*}
\frac{\partial}{\partial t} (\rho \tilde{k}) + \frac{\partial}{\partial x_i} (\rho \tilde{k} \tilde{u}_i) &= \frac{\partial}{\partial x_i} \left( \alpha_k \mu_{eff} \frac{\partial \tilde{k}}{\partial x_j} \right) + G_k + G_b - \tilde{p} \tilde{e} - Y_M + S_k \quad (3.9) \\
\frac{\partial}{\partial t} (\rho \tilde{\epsilon}) + \frac{\partial}{\partial x_i} (\rho \tilde{\epsilon} \tilde{u}_i) &= \frac{\partial}{\partial x_j} \left( \alpha_\epsilon \mu_{eff} \frac{\partial \tilde{\epsilon}}{\partial x_j} \right) + C_{1\epsilon} \tilde{\epsilon} \left( G_k + C_2 \epsilon G_b \right) - C_{2\epsilon} p \tilde{\epsilon}^2 - \epsilon - S_\epsilon \quad (3.10)
\end{align*}
\]

where \( G_k \) represents the generation of turbulent kinetic energy (tke) due to the mean velocity gradients, \( G_b \) is the generation of tke due to buoyancy, \( Y_M \) is the contribution of the fluctuation dilation in compressible turbulence to the overall dissipation rate, \( \alpha_k \) and \( \alpha_\epsilon \) are the inverse effective Prandtl number for \( k \) and \( \epsilon \), respectively and \( S_k \) and \( S_\epsilon \) are user-defined source terms.
3.3 Species Transport

To model the mixing and transport of concentrations of different chemical species in the problem studied here, such as the steam and cyclohexane vapor, a species transport model is used. When solving the conservation equations for chemical species, the local mass fraction of each species, $Y_i$, is calculated from the solution of a convection-diffusion equation for the $i$th species [45]. The conservation equation is described in the general form:

$$\frac{\partial}{\partial t}(\tilde{\rho}Y_i) + \nabla \cdot (\tilde{\rho}\tilde{v}Y_i) = -\nabla \cdot \left[ \sum_{i=1}^{n} h_i \tilde{J}_i \right]$$

(3.11)

where $J_i$ is the diffusion flux of species $n$ and is given by:

$$\tilde{J}_i = -\left( pD_{n,m} + \frac{\mu_t}{Sc_t} \right) \nabla \tilde{Y}_i - D_{T,n} \frac{\nabla T}{T}$$

(3.12)

where $Sc_t$ is the turbulent Schmidt number, which is set to 0.7 and was found to be appropriate for the circumstances in this simulation. For turbulent flows, the mass diffusion is influenced mostly by the turbulent transport. This is determined by the turbulent Schmidt number, which is the ratio of momentum and mass diffusivity [46], [47], [48], [49]. The turbulent Schmidt number is relatively insensitive to the molecular fluid properties, and thus there is little reason to alter the default value of $Sc_t = 0.7$ for any species [45]. $D_{n,m}$ is the mass diffusion coefficient for species $i$ in the mixture, $\mu_t$ is the turbulent viscosity, and $D_T$ is the thermal (Soret) diffusion coefficient. The thermal diffusion is enabled here, which causes heavy molecules to diffuse less rapidly and light molecules to diffuse more rapidly. The thermal (Soret) diffusion coefficient
is computed by applying kinetic theory using the empirically-based expression:

\[
D_{T,n} = -2.59 \times 10^{-7} T^{0.659} \left[ \frac{M_{w,n}^{0.511} X_i}{\sum_{i=1}^{N} M_{w,n}^{0.511} X_n} - Y_n \right] \left[ \frac{\sum_{n=1}^{N} M_{w,n}^{0.511} X_n}{\sum_{n=1}^{N} M_{w,n}^{0.489} X_n} \right],
\]  

(3.13)

where \( M_{w,n} \) is the molecular weight of species \( n \), \( Y_n \) is the mass fraction of species \( n \), and \( X_n \) is the mole fraction of species \( i \) \[45\]. The diffusion flux term on the right hand side of equation 3.11 includes the enthalpy, which accounts for the treatment of species transport in compressible flow. When modeling the secondary phase particles, the degree of coupling between the continuous and discrete phase in terms of the Stokes number, \( St \), needs to be determined. Stokes number is the ratio of relaxation time of particles to hydrodynamic time. It is defined as:

\[
St = \frac{2}{9} \left( \frac{d_p}{L} \right)^2 \frac{Re}{R}
\]

(3.14)

where \( d_p \) is the average particle diameter, \( L \) is the hydrodynamic length scale (the size of the computational domain), \( R \) is defined as follows:

\[
R = \frac{2\rho_f}{2\rho_f + \rho_p}
\]

(3.15)

where \( \rho_f \) is the carrier fluid density, and \( \rho_p \) is the particle density. When the Stokes number is small, i.e. \( St \ll 1 \), the particles are not affected by the fluid flow \[26\]. This means the particles will pass through the continuous phase as opposed to interacting with it, hence the trajectories of the particles remain relatively unchanged. When the Stokes number is large, i.e. \( St \gg 1 \), the particles will interact with the continuous phase, which indicates the trajectories change much more and particle break up will be more significant. The case modeled here has a relatively high Stokes number.
3.4 Multiphase Fluid Flows

When simulating multiphase flows, the Weber number is an important parameter. It’s has been determined that the Weber number is $We = 254$ for the given cement and steam flow. Other important factors to account for with dispersed droplet flow are the characteristic breakup time and the stable/critical droplet diameter. According to Piltch and Ereman [50], the characteristic breakup time is the time it takes for droplets to stop shedding residual child droplets, or when a stable diameter is reached where the surface tension forces overpower other outside hydrodynamic forces. The characteristic breakup time $t$ is defined as:

$$ t = \frac{TD}{U_{rel}^2 \sqrt{\frac{\rho_f}{\rho_d}}} \quad (3.16) $$

where, $U_{rel}$ is the relative velocity between the continuous phase and droplets, $\rho_f$ is the continuous fluid density, $\rho_d$ is the droplet density, and $T$ is the dimensionless time characteristic of droplet breakup due to R-T and K-H instabilities. The dimensionless time characteristic has been correlated by Piltch and Erdman’s finding to correspond to the various Weber number categories. Given the Weber number in this case within the sheet stripping regime, the dimensionless time is:

$$ T = 14.1(We - 12)^{-0.25} \quad (3.17) $$

For our case, the dimensionless time is $T = 3.75$, which gives our characteristic breakup time a maximum value of $t = 0.0024$ seconds. This indicates that when
simulating the primary droplet breakup, the droplets need, at minimum, 2.4 milliseconds before reaching a stable diameter. This was taken into consideration when determining an appropriate time to simulate the primary cement sheet breakup.

Additionally, the stable droplet diameter is calculated using the equation presented by Piltch and Erdman:

$$d_{st} = W e \frac{\sigma}{\rho_d U_{rel}^2}$$  \hspace{1cm} (3.18)

Here, a minimum stable diameter is calculated to be 143 micrometers in diameter.

3.5 Volume of Fluid Interface Tracking Method

To perform a simulation showing the primary breakup of the cement injection within the G1 contactor, a separate model is used. In this model, the continuous steam phase is already in motion flowing through the contactor’s main body, and the cement is added in as a liquid. As the liquid enters the contactor, it is quickly sheared apart into droplets. Since the interface between the gas and cement phase is not disperse, but rather a stratified flow where the immiscible fluids are separated by a clearly defined interface, an optimized version of the Volume of Fluid (VOF) interface tracking method is used. This VOF method, introduced by Hirt and Nichols [51, 52], uses a reconstructed interface geometry using a piece-wise constant/”stair-stepped” approximation to calculate the phase volume per computational cell [53]. The VOF
method has been heavily used in research for droplet formation applications [54],
among other cases involving complex multiphase flows.

3.5.1 Coupled Level Set Volume of Fluid

An improved VOF method, called the Coupled Level Set Volume of Fluid (CLSVOF),
was extended by Sussman and Puckett [28]. This method combines the Level-Set (LS)
method, first introduced by Osher and Sethian [55], and the VOF tracking method.
The LS method captures the interface very accurately, but has shown issues with
mass conservation. The LS method, when combined with the VOF approach, only
computes the geometric properties at the interface, while the VOF method calculates
the void fraction, making it a more accurate model when modeling flows with sharp
interfaces.

The volume fraction equation tracks the interface between the gas and liquid
phases. A solution is found by solving the continuity equation for the volume fraction
of one of the phases. For the $i^{th}$ phase, the continuity equation to solve the volume
fraction is given as:

$$\frac{1}{\rho_i} \left[ \frac{\partial}{\partial t} (\alpha_i \rho_i) + \nabla \cdot (\alpha_i \rho_i \vec{v}_u) \right] = \sum_{p=1}^{n} (\dot{m}_{pi} - \dot{m}_{ip})$$ (3.19)

Here, the term $\alpha_i$ represents volume fraction of the $i^{th}$ fluid within the cell,
thus $\alpha_i$ is zero when the cell is empty of the $i^{th}$, 1 when the cell is completely full of
the $i^{th}$ fluid, and between 0 and 1 when the cell contains and interface between the

29
\(i^{th}\) fluid and the other fluid. Additionally, \(\dot{m}_{ip}\) is the mass transfer from phase \(i\) to phase \(p\) and \(\dot{m}_{pi}\) is the mass transfer from phase \(p\) to phase \(q\).

Here, a First Order Upwind implicit scheme is used for the time discretization. Given as:

\[
\frac{\alpha_i^{n+1} \rho_i^{n+1}}{\Delta t} V + \sum_f \left( \rho_i^{n+1} U_f^{n+1} \alpha_{i,f}^{n+1} \right) = \left[ \sum_{p=1}^{n} \left( \dot{m}_{pi} - \dot{m}_{ip} \right) \right] V \quad (3.20)
\]

where \(n\) is the index for the previous time step, \(n+1\) is the index for the current time step, \(\alpha_{i,f}\) is the volume fraction of the \(i^{th}\) fluid, \(V\) is the volume of the cell, and \(U_f\) is the volume flux through the face of the cell, based on the normal velocity.

The energy equation, which is shared among phases, is given as:

\[
\frac{\partial}{\partial t} (\rho E) + \nabla \cdot \left( \bar{v} (\rho E + \rho) \right) = \nabla \cdot (k_{eff} \nabla T) \quad (3.21)
\]

where the energy \(E\) and temperature \(T\) are time mass-averaged variables:

\[
E = \frac{\sum_{i=1}^{n} \alpha_i \rho_i E_i}{\sum_{i=1}^{n} \alpha_i \rho_i} \quad (3.22)
\]

where \(E_i\) is based on specific heat of the \(i^{th}\) phase and shared temperature. Also, the density \(\rho\) and the effective thermal conductivity \(k_{eff}\) are shared by the phases.

To account for the level-set function within the CLSVOF method, additional terms are added to the momentum equation. The level-set function \(\phi\) is defined as
a distance to the interface. Thus the interface is the zero level-set \( \phi(x,t) \) and is expressed as \( \Gamma = \{ x | \phi(x,t) = 0 \} \) for a two phase flow. The level-set function is then given as:

\[
\phi(x,t) = \begin{cases} 
+|d| & \text{if } x \in \text{the primary phase} \\
0 & \text{if } x \in \Gamma \\
-|d| & \text{if } x \in \text{the secondary phase}
\end{cases} 
\]  

(3.23)

where \( d \) is the distance from the interface. Also, the normal \( n \) and curvature \( \kappa \) of the interface is needed when considering surface tension, and is estimated as:

\[
\vec{n} = \frac{\nabla \phi}{|\nabla \phi|} \Big|_{\phi=0} 
\]  

(3.24)

\[
\kappa = \nabla \cdot \frac{\nabla \phi}{|\nabla \phi|} \Big|_{\phi=0} 
\]  

(3.25)

The continuity equation for the level-set function within the VOF method (i.e. the CLSVOF method) is written as:

\[
\frac{\partial \phi}{\partial t} + \nabla \cdot (\vec{u} \phi) = 0 
\]  

(3.26)

And the momentum equation can be written as:

\[
\frac{\partial}{\partial t} (\rho \vec{u}) + \nabla \cdot (\rho \vec{u} \vec{u}) = -\nabla p + \nabla \mu \left[ \nabla \vec{u} + (\nabla \vec{u})^T \right] - \sigma \kappa \delta(\phi) \nabla \phi + \rho \vec{g} 
\]  

(3.27)

where \( \delta(\phi) \) is defined as:
\[ \delta(t) = \frac{1 + \cos\left(\frac{\pi \phi}{a}\right)}{2a} \]  

(3.28)

when \(|\phi| < a\) for \(a = 1.5h\), where \(h\) is the grid spacing. For any other values of \(|\phi|\), \(\delta(t) = 0\).

3.6 Discrete Phase Model

3.6.1 Particle Motion

To predict the trajectory of the discrete phase particles, the model used integrates the force balance on the particle, which is written in a Lagrangian reference frame. This force balance equates the particle inertia with the forces acting on the particle, and can be written as:

\[ \frac{d\vec{u}_p}{dt} = F_D(\vec{u} - \vec{u}_p) + \frac{\vec{g}(\rho_p - \rho)}{\rho_p}, \]  

(3.29)

where \(F_D(\vec{u} - \vec{u}_p)\) is the drag force per unit particle mass, given by:

\[ F_D = \frac{18\mu C_D Re}{\rho_p d_p^2 24}, \]  

(3.30)

and \(\vec{u}\) is the continuous phase velocity, \(\vec{u}_p\) is the particle velocity, \(\mu\) is the dynamic viscosity of the fluid, \(\rho\) is the density, \(\rho_p\) is the density of the particle, and \(d_p\) is the particle diameter.

In addition, the simulations use a high-Mach number drag law to account for a particle Mach number greater than 0.4 and particle Reynolds number greater than
20. This drag coefficient is based off of the drag applied to a spherical droplet [56], but incorporates an additional term [57, 58] and can be written as:

\[
C_D = 1 + Kn\left(2.514 + 0.8e^{-0.55/Kn}\right)
\] (3.31)

where \(Kn\) is the Knudsen number and is the ratio of the molecular mean free path in the continuous fluid to the characteristic particle length (here, the particle diameter) [57]. Accounting for the compressibility of an ideal gas, the Knudsen number can be written as [59]:

\[
Kn = \sqrt{\frac{\pi k Ma}{2 Re}}
\] (3.32)

where \(k\) is the ratio of specific heats of the gas and droplet phase.

3.6.2 Multicomponent Mass and Heat Transfer

Using the DPM, reactions of the particles and how they affect the continuous phase can be examined using one of the heat and mass transfer laws in Fluent [60]. The particles in this case may be composed of either pure liquid cyclohexane, pure polymer, or a combination of both. Since this study focuses on particles containing two species, the multicomponent particle heat and mass transfer conditions are used. The multicomponent droplet vaporization rate is calculated as the sum of the vaporization rates of the individual components, where for component \(i\), the particle mass \(m\) is the sum of the masses of the components, ie. \(m = \sum_i m_i\). Using a convection/diffusion controlled vaporization model, the vaporization rate is solved as [45]:

33
\[ \frac{dm_i}{dt} = A_p k_{c,i} \rho_g \ln(1 + B_{m,i}) \] (3.33)

where \( m_i \) is the mass of component \( i \) in the droplet, \( k_{c,i} \) is the mass transfer coefficient of component \( i \), \( A_p \) is the droplet surface area, \( \rho_g \) is the density of the gas phase, and \( B_{m,i} \) is the Spalding mass number for species \( i \), which is given as:

\[ B_m = \frac{Y_{i,s} - Y_{i,g}}{1 - Y_{i,s}} \] (3.34)

where \( Y_{i,s} \) is the vapor mass fraction at the surface, and \( Y_{i,g} \) is the vapor mass fraction in the gas phase. To account for boiling due to high pressures, the boiling rate equation is solved, and is given as [45]:

\[ \frac{dm_i}{dt} = x_i \frac{\pi d_p k_g}{c_{pg}} (2 + 0.6 Re_d^{1/2} Pr^{1/3}) \ln(1 + B_{T,i}) \] (3.35)

where \( x_i \) is the volume fraction of component \( i \) in the droplet, \( d_p \) is droplet diameter, \( k_g \) is the thermal conductivity of the continuous gas phase, \( c_{pg} \) is the specific heat of the continuous gas phase, and \( B_{T,i} \) is the Spalding heat transfer number for component \( i \), defined as:

\[ B_{T,i} = \frac{c_{pg}(T_g - T_p)}{h_{vap,i}} \] (3.36)

The energy equation for the multicomponent particle is written as [45]:

\[ m_p c_p \frac{dT}{dt} = h A_p (T_g - T_p) + \sum_i \frac{dm_i}{dt} (h_{vap,i}) \] (3.37)
3.6.3 Discrete Random Walk Particle Dispersion

To track the cement particles as they flow through the domain, the stochastic discrete random walk (DRW) model is used [61]. This method uses the velocity fluctuations caused by turbulence to predict the particle trajectory, thus the interaction of the particle with turbulent eddies is simulated [62]. Each eddy is characterized by random velocity fluctuation \((u', v', w')\) and a time scale, \(\tau_e\). For the random velocity fluctuations within the turbulent eddy, these are taken assuming they obey a Gaussian probability distribution such that \(u' = \zeta \sqrt{u'^2}\). Here, \(\zeta\) is a normally distributed random number and the \(\sqrt{u'^2}\) term is the local RMS value of the velocity fluctuation. This RMS value is applied to each velocity flow direction component, and since the kinetic energy of turbulence is known at each point in the flow for the \(k-\epsilon\) turbulence model, the RMS fluctuations can also be defined as \(\sqrt{2k/3}\) assuming isotropy.

The characteristic eddy lifetime is defined as:

\[
\tau_e = 2T_L \tag{3.38}
\]

where \(T_L\) is estimated as \(0.3(k/\epsilon)\) [63].

3.6.4 Two-Way Coupling

To account for the coupling between the gas and particle phase, momentum exchange, heat exchange, and mass exchange are all calculated within the simulation. For any of these quantities, if one is lost or gained from the particle, they are incorporated
into the continuous phase calculations. The momentum exchange equation is given as [45]:

\[
F = \sum \left( \frac{18 \mu C_D Re}{\rho_p d_p^2 24} (u_p - u) \right) m_p \Delta t
\]  

(3.39)

where \( \mu \) is the viscosity of the fluid, \( \rho_p \) is the particle density, \( d_p \) is the particle diameter, \( u_p \) and \( u \) are the particle and fluid velocity, respectively, \( \dot{m}_p \) is the flow rate of the particles, and \( \Delta t \) is the time step. The heat transfer from the discrete particle phase to or from the continuous phase can be computed from [45]:

\[
Q = \frac{\dot{m}_{p,0}}{m_{p,0}} \left[ \left( m_{p,in} - m_{p,out} \right) \left[ -H_{lat,ref} \right] - m_{p,out} \int_{T_{ref}}^{T_{p,out}} c_p dT + m_{p,in} \int_{T_{ref}}^{T_{p,in}} c_p dT \right]
\]  

(3.40)

where \( \dot{m}_{p,0} \) and \( m_{p,0} \) are the initial mass flow rate and mass of the particle, respectively, \( m_{p,in} \) is the mass of the particle on cell entry, \( m_{p,out} \) is the mass of the particle on cell exit, \( c_p \) is the heat capacity of the particle, \( T_{p,in} \) and \( T_{p,out} \) are the temperatures of the particle on cell entry and cell exit, respectively, \( T_{ref} \) is the reference temperature for enthalpy, and \( H_{lat,ref} \) is the latent heat at reference conditions. The mass transfer from the discrete particle phase to the continuous phase is calculated from [45]:

\[
M = \frac{\Delta m_p}{m_{p,0}} \dot{m}_{p,0}
\]  

(3.41)
CHAPTER IV

COMPUTATIONAL DETAILS

4.1 Mesh Details

4.1.1 G1 Contactor: Primary Breakup

Even though the primary breakup is modeled in a shorter section of the contactor, because it is a full 3D coupled level set volume of fluid model, it is computationally expensive. Therefore there is a necessary balance between accuracy and efficiency. The accuracy of the relevant parameters like velocity, temperature, and particle location is highly dependent on the mesh refinement.

Since a droplet cannot be smaller than the mesh element in order to accurately model the interface, the stable diameter of droplets is a key component of mesh size. The stable diameter for this research is calculated to be 143 micrometers, which requires that the maximum mesh length be shorter than that. As a result, the mesh size for models ranges from approximately 2.87 million elements and 3.01 million nodes. Having chosen a hexahedral dominant mesh for accuracy and convergence speed, more than 99% of the elements in all of the models are hexahedral as seen in Figure 4.1.
Figure 4.1: View of dual steam model mesh
4.1.2 G1 Contactor: Secondary Phase Change

The entire contactor length is used in the 3D axisymmetric model to evaluate the secondary phase change and track particle locations. Similar to the mesh for primary breakup, a hexahedral dominant mesh is used to improve accuracy and convergence. For this research, a mesh size of approximately 725,000 elements and 795,000 nodes is used and is shown in Figure 4.2. Earlier research shows, through mesh dependence tests, that this mesh size is sufficient to accurately capture the parameters of interest [5, 6, 4].

4.2 Solution Methods

As described in the problem description, primary breakup is simulated using a coupled level-set volume of fluid (CLSVOF); and secondary phase change is evaluated with a Discrete Phase Model (DPM). The solution methods and parameters associated with each model in ANSYS Fluent are described in the sections that follow.

4.2.1 Coupled Level-Set Volume of Fluid

The CLSVOF model is a transient simulation that simultaneously solves for both the continuous and discrete phases (steam and cement, respectively). Listed below are the schemes used to solve the model based on research and ANSYS guidance [45, 64, 65].

- Pressure-Velocity Coupling: SIMPLE
- Pressure: PRESTO! (PREssure STaggering Option)
Figure 4.2: 3D axisymmetric contactor view of (top) mixing zone and gap region near the end of the contactor (bottom) outlet and divergence region
• Momentum, Density, Energy, Turbulence: First Order Upwind

• Gradient: Least Squares Cell Based

• Level-set Function: First Order Upwind

• Time: First Order Upwind Implicit

• Volume Fraction: Geo-Reconstruct

The Geo-Reconstruct scheme is particularly useful for time-dependent solutions with sharp interface between phases like those in this research.

4.2.2 Discrete Phase Model

Where the CLSVOF model uses First Order Upwind to speed convergence, the DPM simulation benefits of the accuracy of Second Order Upwind discretization. In addition, there are specific parameters associated with particle tracking that require careful consideration. These are generally related to how the model performs coupled calculations between the discrete and continuous phases - when the cement is allowed to impact the steam flow.

One of the major coupling parameters is the number of continuous phase iterations per DPM iteration. For this research, 4 continuous phase iterations occur before the next parcel of particles are injected into the flow. Increasing these iterations can be beneficial when the steam flow convergence is slow for various reasons (e.g. high particle mass loading), but it also becomes computationally expensive. Once the particles are injected, a maximum number of trajectory time steps is set to 42,000 with
a default step length factor of 5. This allows the simulation to abort trajectories for trapped particles, showing those particles then as "incomplete." A general guideline for the time steps is the number of grid cells in the flow direction times the step length factor, which in this case results in the 42,000 steps (5 calculations in each cell) [45]. The time step necessary to progress the DPM injections and solve for particle trajectories is 0.0001 seconds.

All boundaries in the model are set as reflective, except for the outlet. For the purposes of this simulation, the particle interaction with the walls of the contactor is assumed to be completely elastic, meaning the particle retains all of its normal and tangential momentum after the collision [45]. The outlet is set to an escape boundary condition so that it is the only exit for the particles from the contactor except in cases of reverse flow at the inlets.
CHAPTER V

RESULTS

5.1 Primary Breakup

The simulations to measure primary breakup are set up using a CLSVOF model in ANSYS Fluent for a shorter 254 millimeter section of the full 3D contactor. This length has been determined to be sufficient for viewing the initial liquid atomization by previous studies [5] and validated, visual results from this research. The intent is to capture just the initial droplet formation without going through the computational expense of seeing further phase change, which will be modeled in a separate 3D axisymmetric DPM model.

In each case, the transient model is started using a time step of 1e-5 with only steam flowing through the contactor until it reaches a uniform flow at the outlet under the desired conditions. Because the industry partner for this research specified a mass flow rate of 3.9 kg/s and a steam/cement ratio ranging between 0.6 and 1.0, the steam pressure is steadily adjusted until the ratio is achieved. Because, as in previous research, an optimized outcome is the use of as little costly, superheated steam as necessary, each model is tuned to get as close to 0.6 as possible.

Once the cement injection starts, the phase interaction and mass difference
has an immediate impact on the steam flow, steadily dropping the flow rate of the steam at all inlets as more and more cement flows into the contactor. The steam inlet pressure is periodically adjusted upwards until the steam/ratio is restored to the original value and the flow at the outlet becomes reasonably uniform. As a result, the final steam pressure at all of the inlets for each model is 1.41 MPa, leading to steam/cement ratios all within the specified range and within less than 5% of each other.

5.1.1 Particle Diameter Distribution

Because the flow is turbulent, the breakup and outlet conditions are never fully uniform. As such, once the inlet and outlet flow conditions seem to reach a relative equilibrium and uniformity, images of the breakup were monitored for consistency. When the primary breakup became mostly uniform, several different images of the simulation are taken at arbitrary points in time and analyzed using an image analysis code in Matlab. An example of output of the code is shown in Figure 5.1, which provides a visual for how the code isolates individual droplets for measurement.

The diameter distributions are plotted in histogram form where anything over 3000 microns is neglected as being either a part of the initial liquid stream or captured against the wall without separation, leaving the total histogram percentage less than 100%. Because of this limitation, little consideration is afforded for how much cement is sticking to the wall and the impacts on the final polymer crumb quality. Thus the reason for the research that follows in subsequent sections.
A comparison of the minimum, maximum, and average diameters resulting from the CLSVOF results for each geometry is provided in Table 5.1. Considering just diameter initially, the original geometry’s distribution, visually depicted in Figure 5.2, appears favorable with a majority of particles being less than 1000 micron. It also has the smallest droplets created at only 181 micron in diameter. This, in large part, is due to separation that occurs as the cement droplets "shed" from the column of fluid moving along the wall. This primary breakup is different than in any of the other geometries considered and reflects more of a bag-type breakup described earlier.

The second case considered is the geometry that includes the addition of another steam inlet downstream of the cement injection slit. Both steam inlets inject
Table 5.1: Minimum, average, and maximum diameters in microns for each geometry (red italics denote highest values)

<table>
<thead>
<tr>
<th>Geometry</th>
<th>Min</th>
<th>Avg</th>
<th>Max</th>
</tr>
</thead>
<tbody>
<tr>
<td>Original</td>
<td>181</td>
<td>944</td>
<td>1998</td>
</tr>
<tr>
<td>Dual Steam - 45°</td>
<td>338</td>
<td>1285</td>
<td>2448</td>
</tr>
<tr>
<td>Dual Steam - 90°</td>
<td>127</td>
<td>1662</td>
<td>1941</td>
</tr>
<tr>
<td>Reversed Inlets</td>
<td>313</td>
<td>890</td>
<td>1688</td>
</tr>
</tbody>
</table>

Figure 5.2: Upper plane view of Original Geometry CLSVOF results and associated particle diameter distribution from image analysis
superheated steam at a 45° angle in the direction of the contactor flow. This geometry yields a very uniform, column-type breakup where droplets are being sheared from the tip of the liquid cement steam near the center of the annular space as seen in Figure 5.3. However, because the momentum of some of the droplets carries them closer to the wall initially, their velocity slows, allowing droplets from behind to catch up and coalesce. The result is larger particle diameters exiting the outlet. The wavy
pattern of the droplet flow is the result of a slight oscillation that occurs from the phase interaction between the cement and steam flow. In this case, the downstream and upstream inlets continuously over-compensate for one another’s affect on the cement stream.

![Particle Diameter Distribution](image)

Figure 5.4: Lower plane view of Dual Steam - 90° CLSVOF results and associated particle diameter distribution from image analysis

In third case, the downstream steam inlet injects superheated steam normal to the wall of the contactor rather than at a 45° angle as in the previous case. This downstream steam inlet forces the column of cement liquid to become more vertical, which leads to a reduced flowrate of steam coming from the upstream inlet as it
contacts the dense cement mixture. This tension between the two steam flows leads to higher aerodynamic forces, shearing the cement unevenly and nearer the opposite wall. It manifests itself in the image and histogram in Figure 5.4 as a larger number of small diameter particles than in the 45° case but the same coalescence of droplets near the walls.

In the final case, cement is injected behind two opposing steam inlets injecting steam at 45° angles in the direction of the contactor flow as shown in Figure 5.5. Here, the geometry combines the bag-type and column-type breakup regimes previously
considered in the dual steam inlet cases. The effects of gravity on the cement, largely unhindered by the steam ahead of it, pull it to the bottom of the annular space where it begins to accumulate. As the accumulation of cement flows over the inner steam inlet, the superheated steam begins to evaporate cyclohexane and push the column of steam upwards. The counteracting outer steam inlet is also evaporating solvent and forcing the cement downwards. This tension leads to the finer droplets seen in the 90° dual inlet case, while the complimentary flow angles of the steam yield relatively uniform breakup. The same flow oscillation seen in the other geometries also occurs in the reversed inlets geometry, but because the particles are smaller initially, coalescence still results in relatively small diameter particles.

5.1.2 Trapped Cement

The evaluation and optimization of primary and secondary breakup in the polymer devolatilization process leads to highly accurate and insightful results [5, 3, 4, 6]. However, this research suggests that it is equally as important to evaluate that which did not break up, namely the cement that sticks to the contactor walls, reducing steam contact and annular space flow area.

In order to quantify and compare the amount of cement sticking to the walls in each geometry, the volume flowrate of cement near the walls at the outlet is monitored throughout the duration of the CLSVOF simulations. Figure 5.6 shows the inner and outer edges of the outlet that are monitored for cement volume passing through them. Each surface is 3 millimeters in radial width. While the flow is relatively uniform, a
short sampling of the results is measured and plotted in Figure 5.7 for each geometry.

As expected, the plotted results closely align with the still images of the CLSVOF models in the earlier figures. The original geometry has large spikes that depict the sizable liquid structures exiting the outlet as they begin to separate from the larger column of cement adhering to the wall upstream.

The case with the second largest amount of cement near the wall is the geometry with dual steam inlets flowing at 45° angles. The reason for this is not necessarily intuitive when viewing Figure 5.3, which visually appears as though it has the least cement against the wall of all the geometries considered. However, when multiple images are evaluated, the reason becomes more clear; and that reason is the oscillation and coalescence mentioned in the previous section. As the droplets begin to coalesce, the oscillatory pattern of the cement flow is such that the droplets exit the geometry very near or against the edges right at the outlet. The other geometries likely depict less cement sticking to the wall because it adheres to the wall much closer to the inlet and "creeps" along the wall due to the velocity profile.
Figure 5.7: Comparison of Rosin-Rammler particle diameter distributions for each geometry through the annular space. This is further supported by the results from the reversed inlets model where the oscillation is forcing some cement against the wall near the inlet, and the small, coalescing particles moving towards the wall near the outlet are being captured in the monitor as well.

Since the Dual Steam - 90° geometry has a slightly less favorable diameter distribution than the original geometry, and because it has noticeably less cement sticking to the wall – particularly near the inlet – it will be the model used to compare secondary phase change. The results from this comparison should provide insight into the effects of cement adhering to the wall given a neutral or slightly less advantaged particle diameter distribution.
5.2 Secondary Phase Change

The DPM simulations developed in ANSYS Fluent are generated in much the same way as the CLSVOF models. The major geometry difference is that a 3D axisymmetric model of the entire contactor is used in order to measure relevant parameters of the outlet particles prior to entering the coagulation process. A steady DPM model with unsteady particle tracking is utilized to account for both the position tracking and phase change of each particle.

In order to transition from the CLSVOF model to the DPM model, the particle diameters determined from the former are fit to a Rosin-Rammler distribution and inserted as a boundary condition for the injections. A comparison of each geometry’s diameter distribution is provided in Figure 5.8. The simulations are started with only steam to achieve an ideal steam/cement ratio. As expected, the inlet steam pressure of 1.41 MPa used in the CLSVOF models proves to again be the correct pressure for the DPM models as well. After uniform flow is achieved, the particle injection was started. In order to achieve 3.9 kg/s mass flowrate, the injections ranges from 2400 to 2800 particles with each 1e-4 time step. By setting up a monitor to measure cyclohexane vapor leaving the outlet, the simulation shows an immediate spike of cyclohexane vapor at the outlet upon each injection of particles, which can be assumed to be the flash caused by pressure drop in the cement injection and initial contact with the steam.

Immediately after the initial injections, a small amount of particles exit the
As more and more particles enter the contactor, the steam flow begins to steadily drop until it reaches a new equilibrium. In the original contactor, the steam inlet pressure is increased to 2.2 MPa in order to at least maintain a 50% steam/cement ratio. Incremental adjustments beyond this already 93% increase from the original inlet pressure make the model highly susceptible to divergence in the nanogap region depicted in previous DPM mesh figures. Conversely, the dual steam
inlet model does not require an inlet steam pressure adjustment in order to maintain the same 50% steam/cement ratio.

Upon further investigation, the significant increase in steam inlet pressure required in the original geometry is mainly the result of overcrowding in the annular space before the nanogap. In other words, the particles in the space are oriented in such a way that there is insufficient contact with the superheated steam; and as more and more of these particles are injected with minimal cyclohexane evaporation, the steam has less and less room to flow through the contactor, requiring added inlet pressure. Eventually the model reaches some equilibrium when there is sufficient steam flow around or over the particles to begin to pushing them through the nanogap. This closely follows what can be seen in the CSLVOF model for the original geometry where the cement begins sticking to the wall immediately after injection.

For both models, the particle tracking is set to 42,000 iterations, which is a function of both the step length and number of elements along the flow path of the fluid. If after 42,000 iterations of the trajectory calculations, the particle cannot be properly placed, the model assumes that the particle is stuck inside a vortex or eddy, and it terminates the particle as "incomplete," removing it from the simulation. For large numbers of particles that cannot be placed properly in the flow field, this becomes an increasing computational expense. For the original geometry, by the end of the simulation, more than 13,000 particles are being marked as "incomplete" with each injection of approximately 2400 particles - even after increasing the tracking iterations by an order of magnitude to 420,000. This suggests that many particles
Table 5.2: Comparison of outlet particle parameters

<table>
<thead>
<tr>
<th></th>
<th>Diam (µm)</th>
<th>Res Time (s)</th>
<th>Cyclohexane MF</th>
<th>Temp (K)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Original (Min)</td>
<td>84.7</td>
<td>0.021</td>
<td>0.000</td>
<td>283.7</td>
</tr>
<tr>
<td>Dual Steam (Min)</td>
<td>59.9</td>
<td>0.019</td>
<td>0.000</td>
<td>271.9</td>
</tr>
<tr>
<td>Original (Avg)</td>
<td>727.8</td>
<td>0.046</td>
<td>0.537</td>
<td>322.2</td>
</tr>
<tr>
<td>Dual Steam (Avg)</td>
<td>699.8</td>
<td>0.031</td>
<td>0.484</td>
<td>321.5</td>
</tr>
<tr>
<td>Original (Max)</td>
<td>1687.4</td>
<td>0.069</td>
<td>0.815</td>
<td>474.7</td>
</tr>
<tr>
<td>Dual Steam (Max)</td>
<td>1589.3</td>
<td>0.038</td>
<td>0.768</td>
<td>493.7</td>
</tr>
</tbody>
</table>

are becoming trapped in the contactor and are not proceeding along the flow path in the amount of time commensurate with the continuous phase flow (i.e. steam). Therefore, it requires an injection of approximately 1.4 million particles in the original geometry to see as many particles exit the contactor as are being injected. For the dual steam geometry, no particles are marked as "incomplete," and it only requires 400,000 particles to be injected before the number exiting equals the number injected. The computational difference in expense between the original geometry and the dual steam geometry simulations is the equivalent of going from days to hours.

When the number of particles exiting equal the number being injected and when the flow profiles are relatively uniform, a sample of the particles at the outlet is taken in order to evaluate diameter, cyclohexane mass fraction, residence time, and temperature. The comparison of those results is shown in Table 5.2. From this table
Figure 5.9: Histograms comparing key parameters of the particles at the outlet
and from the corresponding histogram plots of the same parameters in Figure 5.9, there a couple of results that stand out. The results of the original geometry have been validated with experimental data measured in the manufacturing facility of the industrial partner. Because physics are similar for the slight revision in geometry, temporal convergence is assumed to hold true for the new contactor design also [4, 6].

First, while the effects on the minimum and maximum diameters are relatively equal between the models, the average particle diameter in the dual steam geometry is reduced by 30% more than in the original geometry.

Second, a striking result that seems counter-intuitive to the first, is that the maximum particle residence time is 45% lower than in the original geometry. Without the results, it could be errantly assumed that the average and maximum particle diameters would be larger due to less time in contact with the superheated steam and therefore less solvent evaporation. With regard to the former, as mentioned previously, the results clearly show lower minimum, maximum, and average particle diameters than resulted from the original geometry simulation. And as it relates to the cyclohexane mass fraction, Figures 5.12 and 5.13 show that the content is noticeably lower before the converging-diverging region of the contactor due to elevated particle temperatures.

To eliminate the theory that increased particle velocity, resulting from the second steam inlet, causes the reduced residence time, a visual comparison is provided in Figure 5.14. While there may be a very slight elevation in particle velocities in the main mixing area of the contactor, velocities near the outlet are similar, providing
little to support such a significant decrease in residence time.

However, it is through this figure and the others that another theme begins to form, which is likely the reason for the enhanced performance of the dual steam design. At the outer edges of the original contactor, there are darkly colored rows of particles depicting very high levels of cyclohexane mass fraction and exceedingly low values of temperature and velocity. This is increasingly true at the bottom of the contactor where it is likely that large stagnate particles have settled to the bottom of the contactor due to gravity. This is further supported by the image of residence time, where the particles with the longest residence time are coming from the bottom of the contactor. These particles would represent larger, stagnate cement structures that have finally been sheared away from the cement adhering to the wall.

In the dual steam design, this either occurs to a much lesser extent or not at all, because the figures depict a more uniform set of properties. All of these things are the result of mixing with the superheated steam. Where there is insufficient contact with the steam, as in the case of geometries like the original contactor where much of the cement is contacting the wall, then there is poor solvent removal regardless of residence time. When the amount of cement sticking to the wall is reduced, cyclohexane removal is enhanced, and the particles flow through the contactor as intended.

Therefore, this research suggests that optimizing the polymer devolatilization process in steam contactors by only evaluating the quality of the breakup and droplet formation is insufficient if not also paired with an intentional focus on reducing the amount of cement sticking to the wall.
Figure 5.10: Particle diameters tracks for (Top) Original Geometry and (Bottom) Dual Steam Inlets - 90°

Figure 5.11: Particle residence time tracks for (Top) Original Geometry and (Bottom) Dual Steam Inlets - 90°
Figure 5.12: Particle cyclohexane mass fraction tracks for (Top) Original Geometry and (Bottom) Dual Steam Inlets - 90°

Figure 5.13: Particle total temperature tracks for (Top) Original Geometry and (Bottom) Dual Steam Inlets - 90°
Figure 5.14: Particle total temperature tracks for (Top) Original Geometry and (Bottom) Dual Steam Inlets - 90°
CHAPTER VI

CONCLUSION AND FUTURE WORK

6.1 Summary of Results

To appropriately measure results, it is important to compare the findings against the original intent of the research. That purpose, in this study, was to accurately model the polymer devolatization process in a steam contactor and measure the resulting effects of geometry changes on sheet breakup and the amount of cement sticking to the wall. These effects are measured via a parametric study of the outlet particle properties, namely diameter, residence time, and cyclohexane mass fraction. After evaluating the results of the research, the following conclusions can be drawn:

- The interactions between the phases has a significant effect on the operation of the contactor and the effectiveness of the devolatization process. In fact, it is a major consideration in any primary breakup optimization effort. This is apparent even in the problem that this research seeks to resolve where the steam forces the cement against the contactor wall. In each subsequent geometric configuration, the steam pressures require regular adjusting to overcome the phase interaction with the cement and achieve the desired steam/cement ratio.
• The limit placed on the steam/cement ratio adds an element of complexity to optimization options. Through various trial and error simulations, higher steam ratios create finer and much more rapid breakup. However, because of the cost of superheated steam and the industry partner’s desire to conserve it, it is the first condition set in any simulation before evaluating the results, whether positive or negative.

• Even less than ideal geometric arrangements and operating conditions can result in a reasonably ideal diameter distribution. In live production, the original geometry results in polymer crumb quality issues from under- or over-drying of the polymer thought to be caused by cement sticking to the contactor walls. However, when looking at just the primary breakup diameter distribution, it is more favorable than some of the other improved geometries. This supports the purpose of this research and the need to evaluate the effects of the cement that does not break up well, if at all.

• The second steam inlet performs its intended function by reducing the amount of cement on the walls without increasing steam consumption. This allows particles to be fully enveloped by superheated steam, increasing heat transfer and cyclohexane evaporation in the body the contactor prior to the nanogap. The end result is the elimination of the crowding of large, stagnate particles in the annular space near the inlets as well as near the nanogap. Instead, in the revised geometries, most of the particles are continuously progressing through
the contactor and being reduced in size through cyclohexane evaporation, so that they pass more readily into the nanogap prior to arriving. This, and not a particle velocity increase, is the most significant driver for the reduction in overall residence time by nearly 50%.

The last item is really the foremost result of the research and the crux of this study: that even with non-ideal primary breakup diameters, a geometric or operating condition change that reduces the amount of cement sticking to the wall can have a significant effect on final product quality. This conclusion combined with other proven optimization techniques like increasing turbulent mixing and residence time can have profound impacts on the polymer devolatilization process in steam contactors [3, 4, 6].

6.2 Recommendations for Future Work

Based on the results of this research, there are some natural recommendations for future work that arise. The first step would be to complete the DPM simulations for the remaining two geometries presented in this study to develop a full parametric comparison of the effects of diameter distribution and the amount of cement sticking to the wall. This could lead to additional useful conclusions to further optimize the contactor design.

As previously discussed, the particles within the second multiphase simulation likely undergo secondary breakup. Since the first simulation showing primary droplet breakup is restricted to the first 254 millimeters of the contactor domain, a KHRT
secondary breakup model could be enabled within the DPM model as outlined in previous research and below [4]. This would significantly enhance the final output of the DPM model to reflect actual conditions in the manufacturing facility.

There is also very clearly more work to do regarding the optimization of the geometry in order to minimize the amount of cement that sticks to the wall. This research has addressed the benefits of reducing that volume but there is ongoing work necessary to determine the ideal geometry modification to eliminate it if possible.

One such geometry to consider would include the evaluation of coaxial gaseous flow by changing the cement inlet location to inject from the rear of the contactor directly into opposing steam flows (seen in Figure 6.1). Two different variations of this arrangement would be evaluated: (1) steam injection at 45° angles and (2) steam injection normal to contactor walls.
Beyond contactor design changes, there are also several different modeling parameter changes that could be adjusted to improve how cement interacts with the wall. For the CLSVOF model, the walls are given a zero heat flux/generation thermal boundary condition and a standard roughness. Because steam is flowing through the contactor prior to cement being injected, there may be a more accurate way to model the effects of the hot walls on the cement as it initially comes into contact with it. This might eliminate some amount of cement that appears to be sticking to the wall.

Similarly, in the DPM model, all the walls are given a "reflect" boundary condition, which causes the particles that collide with the wall to maintain constant momentum at the same deflecting angle at which they hit. In order to better simulate the "wall stick" of interest, however, a different wall treatment in the boundary conditions may be useful to consider.
BIBLIOGRAPHY


