Examining the Effect of Particle-particle and Fluid-particle Interactions on Mixing and Dispersion of Solids

by

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Particle-fluid and particle-particle interaction play an essential role in many applications: the mixing of solids and liquids, granulation, coating, waste removal, oil production, solid rocket propellants, etc. The viscosity of the fluid can affect the efficiency of this process. For example, a high viscosity fluid needs a higher applied shear such that it is typical that a bladed mixer is used for applications such as the mixing of solid rocket propellant. As an alternative, a Resonant Acoustic Mixer (RAM) boasts several purported advantages over a bladed mixer including shorter mixing time and fewer particle breakages. Nevertheless, the mixing mechanisms and critical operational parameters that affect the mixing performance within a RAM are poorly understood.

In this work, the RAM was evaluated using both experiments and computational modeling. It is found that higher viscosities decrease the mixing performance. Interestingly, size differences between particles are critical, and some cases show segregation.

Another case in which particle-fluid interactions play a vital role is in processes that involve a liquid bridge between particles. We examine the dynamics of a liquid bridge between a sphere and a flat plate being separated from each other. Unlike previous research, we focus on the case where the viscosity of the bridge is lower than that of the external fluid. We develop a lubrication theory-based model that is applicable for the general case of a viscosity mismatch between the bridge fluid and the external fluid. The model predicts that a low viscosity bridge reduces the force as compared to both separation without a liquid bridge or separation with a bridge
of matched viscosity. Experiments show that the length of the bridge at rupture increases as the separation velocity increases. Also, the magnitude of the force reduction is experimentally shown to be more severe at small sphere-plate separations and at large bridge volumes, but the magnitude of the reduction is even larger than that predicted from our model. We propose that the lubricating effect of a thin coating of the low-viscosity bridge fluid that is left behind on the sphere and plate is responsible for this discrepancy.
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Preface

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1.0 Introduction

Particle-fluid and particle-particle interaction play an essential role in many applications: the mixing of solids and liquids, granulation, coating, waste removal, oil production, solid rocket propellants, etc. The viscosity of the fluid can affect the efficiency of this process. Viscosity can be defined in simpler words as the resistance of the fluid to flow. A high viscosity fluid needs a higher applied shear in order to move. One specific application involving high viscous fluid is the mixing of high solid loading propellants. Usually, a blade mixer is used to mix ammonium chloride, aluminum powder, and a viscous binder. A Resonant Acoustic Mixer (RAM) is a recently developed type of mixer that has a couple of purported advantages over a blade mixer: short mixing time and fewer particle breakages, which can be relevant for the final burning property of the mix. Although the RAM could be a great alternative over blade mixers, the mixing mechanism and the operational parameters that affect the mixing performance are not well understood. Most of the reports available about RAM for the mixing of solid propellant fuels are technical papers; for this reason, the limitations of the RAM are not well known for this type of application.

Another case in which particle-fluid interaction plays a vital role is in processes that involve a liquid bridge between particles. Much research exists about the breaking of a liquid bridge in a static flow (air) and moving the particle at a constant rate taking into consideration: particle wettability properties, size, the volume of fluid, and viscosity of the fluid. In contrast, the case for the rupture of a liquid bridge that is less viscous than the surrounding fluid is not well understood, despite the relevance of this arrangement. For example, this type of bridge is common in crystal agglomerations and capillary suspensions, or more generally in cases in which a low viscosity binder is added to a high viscosity continuum phase to create capillary bridges between
particles and produce agglomerates. In these cases, a low viscosity liquid bridge will affect the total force exerted on the particle and gaining a more detailed understanding of these cases will be helpful in describing the rupture mechanisms in this type of liquid bridge.

Computational fluid dynamic (CFD) is a set of numerical methods used to predict the solution of problems involving the motion of fluids. A theoretical approach can provide an exact solution to a straightforward case, but the theoretical approach is too complicated and almost impossible to solve for complex cases. As computing power continues to improve, CFD becomes more accurate and effective and this approach has become one of the primary methods for analyzing complex fluid dynamics [1]. The lattice Boltzmann method is a type of CFD that recovers the Navier-Stokes equations using the Chapman-Enskog expansion [2]. LBM is easy to discretize in a rectangular grid and consists of two computational steps: streaming and collision. In this work, LBM was used to capture the fluid-fluid interaction. The Shan-Chen multicomponent model captures the fluid-fluid cohesion and solid-particle adhesion for the liquid bridge studies.

Similarly, the Discrete Element Method (DEM) is a simulation technique that considers a computational solution for the mechanical interaction between each pair of particles. This method integrates Newton's translational and rotational equations to determine the motion of the particles. The particles forces can include frictional, gravitational, and adhesive forces among others. In this work, DEM is used to capture the particle-particle interaction and is coupled to LBM in order to take into consideration the momentum transport between the fluid and particles.

This dissertation is organized in the following way. In Chapter 2, the background of the computational methods used in this work are described. Chapter 3 shows the validation of the particle-particle and particle-fluid interaction and compares our simulation with experimental results. Chapter 4 the mixing performance of a Resonant Acoustic Mixer was examined both
experimentally and computationally. The limitations of this mixer were identified, and in particular it was noted that segregation toward the bottom is relevant when small particles have a lower concentration in the mixture. The next chapter, Chapter 5, experimentally explores the rupture mechanisms that effect a low viscosity liquid bridge inside a more viscous surrounding fluid. In Chapter 6, a multicomponent LBM-DEM model was developed, tested, and used to investigate the low viscosity liquid bridge rupture. Finally, Chapter 7 discusses our conclusions and the outlook of this work.
2.0 Background

2.1 Lattice Boltzmann Method

The Lattice Boltzmann Method (LBM) was developed from the Lattice Gas Cell Automata models (LGCA). LGCA uses a grid or lattice in which every cell stores a Boolean particle or flag. Each lattice node has a direction vector $e_i$ indicating the moving direction. The method consists of two different steps “streaming” and “collision”. In the “streaming” step, every molecule is copied along the direction vector to the relevant neighboring node. In contrast, when two molecules move in the opposite direction in a single cell, they collide and get redirected using a Boolean operation; this step is called a “collision”. LGCA has a couple of disadvantages: large noise-signal ratio, non-Galilean invariance, and the unphysical dependence of the pressure on the velocity field. [3]

The Lattice Boltzmann Method (LBM) is similar to the LGCA, but is more sophisticated and does not exhibit a large noise-signal ratio. An important change in LBM is the replacement of the Boolean flag by the distribution function $f_i$, which describes the proportion of material at that node with momentum in the direction $i$. The distribution function is the primary variable of the LBM and is used to determine the velocity and density of the fluid domain. The distribution function is the probability of a particle in position $i$ at time $t$ exhibiting a velocity $c$.

2.1.1 Lattice Boltzmann Equation

The Lattice Boltzmann equation is given by:

$$f_i(x + c_i \Delta t, t + \Delta t) - f_i(x + t) = \Omega_i(f(x, t))$$  \hfill (2.1)
where $f_i$ is the distribution function, $\Delta x$ is the lattice spacing, $\Delta t$ is the explicit time step, and $c$ is the lattice speed. The collision operator ($\Omega_i$) determines the rate of change of the distribution function after the collision step. The LBM equation evolves with every time step by collision and streaming processes. The left side of equation (2.1) is the streaming process (convection), which propagates the distribution function to the nearest neighbors. On the right side of the equation (2.1) is the collision process in which the arriving particles from all the nearest neighbors collide and the distribution changes. These two steps are repeated until the end of the simulation.

The macroscopic fluid variables, density $\rho$, and momentum flux $\rho u$ are defined as

$$\rho = \sum_i f_i$$

(2.2)

$$\rho u = \sum_i f_i c_i$$

(2.3)

where $u$ is the velocity, and both are calculated at each lattice node using the distribution function. The domain of the distribution function is dependent on the number lattice arrangement $i$.

2.1.2 Lattice Arrangement and Spatial Discretization

In LBM, the common terminology used for the dimension of the problem and the number velocities is $DnQm$, in which $n$ is the dimension number (2 for 2-D and 3 for 3-D) and $m$ is the number of lattice velocities. A minimum of lattice symmetry and equally-spaced lattice is necessary to recover the Navier-Stokes equation and to ensure isotropy of the velocity tensors [4-]
The type of LBM complexity needed depends on the application and geometry of the system. The most popular 2D model is D2Q9. There are three different models in three dimensions: D3Q15, D3Q19, and D3Q27. The number of the velocity vector is important and can affect the model results and the computational time. The D3Q19 is more accurate in comparison to the D3Q15 that is known to have numerical instability [6]. In contrast, D3Q27 requires 27 velocities per node, which increases the number of grids needed to discretize the physical model and the number of floating points. In general, D3Q19 is a middle point with good performance and the least computational expense [5, 7]. In this work, D3Q19 was employed in our model for all the computational work. The components of the lattice velocities for the D3Q19 in a matrix form is given by:

\[
c_i = c \begin{bmatrix}
1 & 1 & 1 & 1 & 0 & 0 & 0 & 0 & 1 \\
0 & 0 & 0 & 0 & 1 & 1 & 1 & 1 & 0 \\
0 & 0 & 0 & 0 & 0 & 1 & 1 & 1 & 1 \\
0 & 0 & 0 & 0 & 0 & 0 & 1 & 1 & 1 \\
0 & 0 & 0 & 0 & 0 & 0 & 0 & 0 & 0
\end{bmatrix}
\]  

(2.4)

2.1.3 Collision Operator

The collision operator, \( \Omega \), models the collision between fluid particles in a cell and can be described as the rate of change of the probability distribution function and is presented as

\[
\frac{df}{dt} = \Omega(f)
\]  

(2.5)

Equation 2.5 is hard to solve analytically, and it is impossible to compute an exact solution. In 1992 a simple approximation was suggested by Bhatnagar, Gross, and Krook (BGK) [8]. In this approximation, a single relaxation time is used to reduce the collision operator and make it easier
to use in computational models. This approximation follows the idea that the distribution function
wants to achieve an equilibrium distribution function \( f_{i}^{eq} \), that is defined as

\[
 f_{i}^{eq} = w_{i} \rho \left( 1 - \frac{3}{2c^{2}} u^{2} + \frac{3}{c^{2}} (c_{i} \cdot u) + \frac{9}{2c^{2}} (c_{i} \cdot u)^{2} \right)
\]  

(2.6)

where \( w_{i} \) is the weight factor of the velocities and for D3Q19 geometry are: \( w_{0} = \frac{1}{3} \), \( w_{7-6} = \frac{1}{18} \) and \( w_{7-18} = \frac{1}{36} \), and \( c \) is the basic speed of the lattice (\( \Delta x/\Delta t \)) usually equal to 1. The BGK collision operator is written as,

\[
 \Omega_{i} (f(x,t)) = - \frac{\Delta t}{\tau} (f_{i}(x,t) - f_{i}^{eq} (\rho, u))
\]  

(2.7)

where \( \tau \) is the relaxation time constant and is related to the viscosity (\( \eta \)) of the fluid \( \tau = \frac{1}{2}(1 + 6\eta) \) [5].

After substituting the collision operator in 2.1 with equation 2.7, we get the LBM equation that is applied in this work,

\[
 f_{i}(x + c_{i}\Delta t, t + \Delta t) = f_{i}(x, t) - \frac{\Delta t}{\tau} (f_{i}(x, t) - f_{i}^{eq} (\rho, u)).
\]  

(2.8)

Equation 2.8 represents the lattice Boltzmann equation and is unconditionally stable while at the same time capturing the fluid behavior correctly. The main limitations of this method are a restriction to lower Mach number (\( \frac{u}{c_s} \)), where \( c_s = \frac{1}{\sqrt{3}} \) and is the speed of sound in LBM. The other limitation is a proper choice of the relaxation time (\( \tau > 0.5 \), which is needed to have a positive viscosity).

2.1.4 Boundary Conditions

Before doing any calculation, boundary conditions are necessary and are crucial to accurately capturing the fluid behavior. There exist several different methods suggested for solid-
fluid boundary conditions that include bounce-back [9], link bounce-back [10-11], and immersed boundary [12]. Most of them can capture the fluid-solid behavior accurately and almost at the same resolution (computational cost). In this work, we only discuss the following conditions: bounce-back, periodic boundary, and immersed boundary conditions. Other more complicated boundary conditions are well documented by A. A. Mohamed [13], Sukop & Thorne [14], Zhou et al. [15]. The boundary condition applied in our model is the immersed boundary condition coupled with the DEM to capture the momentum transport between solid and fluid. This method allows the inclusion in LBM of a large number of moving boundaries (including walls and particles), retaining the LBM two-step process (streaming and collision) by adding an additional term to the collision operator ($\Omega^2$).[16]

2.1.4.1 Periodic Boundaries

Periodic boundary conditions are the simplest to apply, in which “periodic” means that the system is going to connect or wrap itself at the edges. As an example, the node first node in a 3-D arrangement (x(0),0,0) is the neighbor of the last node in the x-direction (x(last),0,0). Therefore, these nodes interact with each other (streaming and collision). Full periodic boundaries can be useful to capturing fluid-fluid interaction only, and in this work, we used periodic boundaries to study the interactions between a two-component fluid system. This method is usually used in combination with bounce-back boundaries (in order to capture fluid-solid or fluid-particle interactions). One typical example is the simulation of a slit wall, where the walls are modeled as bounce-back boundaries, and the open side of the slit uses periodic boundaries with a given velocity.
2.1.4.2 Bounce-Back

Bounce-back boundaries have been used to model solid stationary walls, non-slip conditions, and moving boundary walls and flow over obstacles. Bounce-back boundaries have different variants. A static wall is simple; when there is an incoming particle toward the solid boundary, the particle bounces back into the fluid domain. The most used method locates the wall at half the distance from the lattice sites. Figure 1 shows the process, the distribution functions prior to the bounce back are known from values of $f_7$, $f_4$, and $f_8$. During the bounce back it is assumed that the distribution function hits the wall and bounces back in the opposite direction. In that case, after the bounce back, the distribution functions are given as $f_5 = f_7$, $f_2 = f_4$ and $f_6 = f_8$. This boundary condition is first-order accurate and generates good results with non-moving boundaries; however, with moving boundaries, the position of the solid needs to be readjusted, which can cause a force fluctuation [10-11]. Thus, a modified condition must be used instead.

Figure 1. Bounce-back at the bottom wall for Q2D9.
2.1.4.3 Immersed Moving Boundary Method

The immersed moving boundary condition was first proposed by Noble and Torzynski [16] and retains all the advantages of the Lattice Boltzmann Method (collision step only depends on the local node and the linear streaming step). This method allows the inclusion in LBM of a large number of moving boundaries (including walls and particles), retaining the LBM two-step process (streaming and collision) by adding an additional term to the collision operator ($\Omega^S_i$). This additional term takes into consideration the proportion of the nodal cell that is covered by the solid. Therefore, it improves the calculation of the forces between a solid boundary and the resultant distribution function.

Adding the additional term $\Omega^S_i$ to the collision step and including the body force in the equation (2.9) the resulting equation is

$$f_i(x + c_i\Delta t, t + \Delta t) = f_i(x, t) - [1 - B_n] \frac{\Delta t}{\tau} \left( f_i(x, t) - f_i^{eq}(\rho, u) \right) + B_n \Omega^S_i + [1 - B_n] \frac{G c_i}{K c^2}$$  \hspace{1cm} (2.9)

where $B_n = \sum B(\epsilon_s, \tau)$ is the weighting factor, the sum is over all the contributions of the obstacles (solids), and $\epsilon_s$ is the volume fraction of each solid particle that are inside the volume of the neighbor’s nodes. The limits of $B_n= 0$ implies that it is a pure fluid node, and $B_n = 1$ means that it is a pure solid node.

Two forms of the additional collision term $\Omega^S_i$ were presented by Noble and Torzynski [16]. The first one is based on the bounce-back of the nonequilibrium part of the distribution function that is given by

$$\Omega^S_i = [f_{i'}(x, t) - f_i^{eq}(\rho, u)] - [f_i(x, t) - f_i^{eq}(\rho, u_s)] \hspace{1cm} (2.10)$$

where $f_{i'}$ is the value of the distribution function after the bounce back is applied (as explained above), and $u_s$ is the velocity of the solid, which for a moving wall is known and for
particles is determined with DEM taking into consideration the translational and rotational velocity.

The second form of the additional collision operator is a superposition method. In this method, the equilibrium distribution function is evaluated at the solid velocity, and a term is added, considering the deviation of the distribution function from the equilibrium distribution function. This second form is given by

$$\Omega_i^s = f_i^{eq}(\rho, u_s) - f_i(x, t) + \left[1 - \frac{\Delta t}{\tau}\right] \left[f_i(x, t) - f_i^{eq}(\rho, u)\right]. \tag{2.11}$$

The first form of the additional collision factor requires a simple weighting factor equal to the solid coverage ratio ($B_n = \sum \epsilon_s$). For the second form of the additional collision factor, a more complex weighting factor is needed and is given by

$$B_n(\epsilon_s, \tau) = \frac{\epsilon_s \left(\frac{\tau}{\Delta t} - 0.5\right)}{(1 - \epsilon_s) + \left(\frac{\tau}{\Delta t} - 0.5\right)}. \tag{2.12}$$

This weighting factor is dependent on viscosity, and the relaxation parameter was only tested with a $\tau/\Delta t$ between 0.6 and 1.0. The results show an excellent approximation of the fluid-solid interactions around a cylinder. [16, 3]

Now that the force exerted by the solids in the fluid is calculated, the total hydrodynamic forces affecting the solid can be calculated by summing the change in momentum with the additional collision operator over all the lattice directions at each node, all the fluid boundary, and all solid nodes.

$$F_p = \sum_n B_n \left(\sum_i \Omega_i^s c_i\right). \tag{2.13}$$

Using the same approach, the hydrodynamic torque can be determined by,
\[ T_p = \sum_n \left[ (x_n - X_p) \times B_n \sum_i (\Omega_i^2 c_i) \right], \]  

(2.14)

where \( x_n - X_p \) is the vector coming from the center of rotation to the coupled node. This torque is added to the torque calculated in the DEM method and captures the interaction between the fluid nodes and the solid nodes.

The immersed moving boundary method is applied in our model to coupled LBM with DEM. Because we are interested in investigating the interaction of solids and liquid with moving walls and particles, and this method can capture these interactions accurately.

### 2.2 Discrete Element Method (DEM)

The particle interactions are captured with the Discrete Element Method (DEM). In this method, the bulk flow of the particles is determined by integrating all the forces between individual’s pairs of particles. In reality, macroscopic granular material deforms under stress; these deformations are too complicated to model and predict [17]. Instead, the forces between particles are evaluated, taking into consideration the deformation of the particles as an overlap between the pair of (undeformed) particles [18].

The motion of each particle is determined with the integration of Newton’s equation for translational and rotational motion, Eq. 2.15 and 2.16:

\[ m_i \frac{d^2}{dt^2} r_i = f_{ii} + m_i g \]  

(2.15)
\[
I_i \frac{d}{dt} \omega_i = t_i
\]

where \(m_i\) is the mass of particle \(i\), \(\mathbf{r}_i\) is the position, \(\mathbf{g}\) is the gravity vector, \(I_i\) is the moment of inertia of the particle, \(\mathbf{\omega}_i\) is the angular velocity and \(\mathbf{t}_i = \sum (\mathbf{l} \times \mathbf{F}_t)\) where \(\mathbf{F}_t\) is the tangential forces, and \(\mathbf{l}\) is the vector between the center of particles \(i\) and the tangential forces (represented in Figure 2). The total forces \(\mathbf{f}_{ii}\) includes the normal and tangential forces between particles and the coupling force for momentum transport between particles and the fluid (immersed boundary condition).

2.2.1 Normal Force

The normal force is modeled using the work of Thornton [18], in which the particles are considered to be an elastic-plastic material. At elastic loading, the normal force \(\mathbf{F}_n\) is given by

\[
\mathbf{F}_n = k_n \alpha^{3/2},
\]

where \(\alpha\) is the overlap (deformation) of the particles given by \(\alpha = (R_1 + R_2 - D_{12})\). \(R\) is the particle radius, \(D_{12}\) is the distance between the particles center and \(k_n\) is the normal force constant determined from Hertz theory [19]. The normal force constant is a function of the elastic properties (\(E_i\) Young’s modulus and \(\nu_i\) Poisson ratio) and is defined by,

\[
k_n = \frac{4}{3} \frac{1}{E^*} \sqrt{R^*}.
\]

In equation 2.18

\[
\frac{1}{E^*} = \frac{1}{E_1} + \frac{1}{E_2} - \nu_1^2
\]

\[
\frac{1}{R^*} = \frac{1}{R_1} + \frac{1}{R_2} - \nu_2^2
\]
If the impact velocity is large enough, the normal force exceeds a yield force \( F_y \), and further increases in the force are calculated using the linear expression

\[
F_n = F_y + k_y (\alpha - \alpha_y). \tag{2.21}
\]

The plastic stiffness \( k_y \) is related to the yield force by the expression

\[
k_y = \left(\frac{3}{2}\right) F_y / \alpha_y k_y,
\]

where \( \alpha_y \) is the overlap or deformation at the point of yield. Before the yield force limit, the uploading is purely elastic, while after the yield force limit is reached the unloading is given by

\[
F_n = F_{\text{max}} - k_n \sqrt{R} (\alpha_{\text{max}} - \alpha)^{3/2}, \tag{2.22}
\]

where \( F_{\text{max}} \) is the maximum force attained during the contact, \( \alpha_{\text{max}} \) is the maximum overlap and \( R \) is the ratio between the new contact curvature due to plastic deformation \( (R') \) and the radius before the elastic deformation of the two particles \( (R^*) \). This ratio can be obtained theoretically as a function of maximum force and yield force and is given

\[
\bar{R} = \frac{R'}{R^*} = \frac{F_y}{F_{\text{max}}} \left(\frac{2F_{\text{max}} + F_y}{3F_y}\right)^{3/2}. \tag{2.23}
\]
2.2.2 Tangential Force

Walton and Braun derived the tangential or frictional force used in this work. [20] A detailed derivation and application of these equations are described by Walton [21]. Here, only a general overview is included.

In this method, the tangential displacement parallel and perpendicular to the friction force are calculated separately. Both contributions are combined vectorially and compared to the Amenton’s Law limit \( F_t \leq \mu F_n \), where \( \mu \) is the coefficient of sliding friction). Taking into
consideration the Amonton’s Law limit and to mimic an annular region of micro-slip at the edge of the contact, the frictional stiffness is given by:

\[ k_t = k_{t_0} \left( 1 - \frac{F_t^* - F_t}{\mu F_n - F_t} \right) \gamma, \quad \text{for increasing } F_t \]

\[ k_t = k_{t_0} \left( 1 - \frac{F_t^* - F_t}{\mu F_n + F_t} \right) \gamma, \quad \text{for decreasing } F_t \]

(2.24)

(2.25)

when the total tangential force increases, equation 2.24 is used, and when the total tangential force decreases, equation 2.25 is used instead. In these equations, \( F_t^* \) is the value of \( F_t \) when the direction of tangential slip changes from increasing to decreasing or sideways, and \( \gamma \) is a constant that is typically set equal to 1/3 to resemble Mindlin’s elastic frictional theory [22].

The initial tangential stiffness \( k_{t_0} \) is a function of the Hertzian normal stiffness and is given by

\[ k_{t_0} = \frac{k_n(1-v)}{1-v/2} \]

(2.26)

The new friction force parallel to the old value is calculated using the magnitude of the old friction force and the increment in tangential force due to displacement

\[ F_\parallel = F_t + k_t \Delta s_\parallel, \]

(2.27)

where \( \Delta s_\parallel \) is the displacement parallel to the old friction force. The displacement can be determined from the component of velocity parallel to the friction force, where \( \Delta s_\parallel = v_\parallel dt \) (\( v_\parallel \) is the parallel velocity and \( dt \) is the time step).

Similarly, the perpendicular displacement can be determined (\( \Delta s_\perp \)) and is used to calculate the perpendicular part of the tangential force,

\[ F_\perp = k_{t_0} \Delta s_\perp \]

(2.28)
The new tentative value of the total tangential force can be calculated by adding the two vectors $\mathbf{F}_\parallel$ and $\mathbf{F}_\perp$,

$$F_t = F_\perp + F_\parallel$$  \hspace{1cm} (2.29)

The total tangential force is then attained by comparing with the Amonton’s Law limit ($F_t \leq \mu F_n$), and in the case that the tentative value exceeds this limit, the tangential force is uniformly scaled to be equal to $\mu F_n$ (the limit).
3.0 Code Validation

We need to validate our methodology before using our LBM-DEM model to investigate complex fluid-solid interactions. Here, we applied our model to analyze two different cases: sedimentation and the rebound of a single sphere in a viscous fluid.

3.1 Sedimentation of a Single Sphere

Single sphere sedimentation was simulated with different values of viscosity and fluid density. The specific values are summarized in Table 1.

<table>
<thead>
<tr>
<th>Case</th>
<th>(p_f) [kg/m^3]</th>
<th>(\mu_f) [Ns/m^2]</th>
</tr>
</thead>
<tbody>
<tr>
<td>Case 1</td>
<td>970</td>
<td>373</td>
</tr>
<tr>
<td>Case 2</td>
<td>965</td>
<td>212</td>
</tr>
<tr>
<td>Case 3</td>
<td>962</td>
<td>113</td>
</tr>
<tr>
<td>Case 4</td>
<td>960</td>
<td>58</td>
</tr>
</tbody>
</table>

The sphere simulated is a Nylon bearing with a diameter of \(d_p = 15\) mm and a density \(\rho_p = 1120\) kg/m^3 in order to match experimental values available within the literature. The particle was released in a container with the following dimensions 100 mm of width, 100 mm of depth, and 160 mm of height. In the simulation, the particle is released at 120 mm (Figure 3) from the bottom of the container, and the only force acting in the sphere is the gravity force (mg). The simulation grid used was \(N_x=100, N_y=160\) and \(N_z=100\) nodes and a relaxation parameter \(\tau = 0.75\).
Figure 3. Simulation set up for sedimentation of a single sphere at 120 mm.

The position of the particles for the four cases is tracked with respect to time and plotted in Figure 4. These results were compared with the experimental data taken from Cate et al. [23] (cross symbols), in which the position of the particles was measured using particle image velocimetry. The results show that our model determines the particle position with respect to time accurately. Therefore, matching the fluid-particle interaction for all four fluids proves that the model can deal with moving particles problems and fluids with different viscosities.
3.2 The Rebound of a Sphere in a Viscous Fluid

The Stokes number is a dimensionless number that represents the ratio between the characteristic time of the particle motion and the characteristic time of the fluid (relaxation), taking into consideration the viscous forces and is given by

\[ St = \frac{\rho_p w_p d}{9 \rho v}, \]  

(3.1)
where $\rho_p$ is the particle density, $\rho$ is the fluid density, $w_p$ is the particle velocity, and $\nu$ is the kinematic viscosity. It has been reported that when $St > 10$, the spherical particles will rebound from a wall [24-25]. The rebound of a sphere in a viscous fluid was investigated by Li et al. [26], and we reproduced these experiments with our model. The experiments were done in a rectangular glass tank filled with a solution of glycerol and water with a kinematic viscosity of $4.173 \times 10^{-5}$ m$^2$/sec and a density of $\rho = 1203$ kg/m$^3$. A steel sphere with a diameter of 9.5 mm and a density of $\rho_p = 7780$ kg/m$^3$ and seven release heights were used. As the LBM method has a Mach Number (Ma) limit, in which a Mach number greater than 0.1 creates numerical instabilities, here we only report on two height releases with our model $h_r = 5.5$ mm and $h_r = 10.5$ mm (since the other conditions result in a Mach number > 0.1).

In the simulations, the young modulus $E$ was set equal to 200 GPA and the Poisson’s ratio to 0.33 to match the sphere's physical properties. The results obtained in Figure 5 show the position of the sphere after the rebound, the line is the simulation result, and the blue dots are the experimental results. Our model predicted the position of the sphere with reasonable accuracy. In Table 2, the result for the Reynold's number ($Re = \frac{\rho v d}{\eta}$) shows a good agreement between the experimental result and the computational result. Therefore, the sphere's velocity before the rebound matches the experimental results. In this table, we also compare the result reported by Lee et al. [24] for the $St$ number, and the values are almost the same. These demonstrate that our model can accurately capture the collision process of a sphere and walls in a viscous fluid.
Table 2. Comparison of the Reynolds [26] and Stokes number [24] with our computational result.

<table>
<thead>
<tr>
<th>Height (mm)</th>
<th>Re (Exp.) [26]</th>
<th>St (Sim.) [24]</th>
<th>Re</th>
<th>St</th>
</tr>
</thead>
<tbody>
<tr>
<td>5.5</td>
<td>53</td>
<td>38</td>
<td>55.5</td>
<td>39.6</td>
</tr>
<tr>
<td>10.5</td>
<td>75</td>
<td>53</td>
<td>73.8</td>
<td>53</td>
</tr>
</tbody>
</table>

Figure 5. Position of the sphere as a function of time A) at 10.5mm and B) at 5.5 mm.
4.0 The Mixing Performance of a Resonant Acoustic Mixer with High Solid Loading and a High Viscous Fluid Experimentally and Computationally

Composite solid propellants typically consist of a multi-modal mixture of ammonium perchloride (AP) as an oxidizer, aluminum powder as fuel, and hydroxyl-terminated polybutadiene (HTPB) as a binder. These propellants are mainly used in rocket propulsion and have a high solid loading [27-29]. The physical and burning properties of the propellant fuel are impacted by the ultimate properties of the mixture. Some mixture properties that affect the burning rate of the fuel include the particle size distribution, particle shape, total solid loading, and the composition of AP and aluminum powder content [31-33]. The mixing parameters that can affect the mixture quality and therefore the physical and burning properties include the type of mixer, mixing time, and the mixing speed [34]. Typically, a vertical blade mixer is used to mix composite solid propellant fuel [35]. In this type of mixer, the mixing time is often more than an hour. In this work, we examine the performance of an alternative mixing device, the resonant acoustic mixer (RAM).

RAM is a non-contact mixing technology that uses a low-frequency acoustic field to promote mixing. [36] This new mixing technology has been shown to have several advantages when used for the mixing of powders, gases, liquids, high viscosity materials, or complex and multiphase systems. [36-40] The principle operating mode of RAM, as shown in Figure 6, is to generate numerous micro-mixing zones throughout the whole system by shaking the mixing vessel vertically. This simple mixing characteristic has made the RAM mixer attractive for study in multiple applications: mixing of pharmaceutical powder [38], increasing the mass transfer of oxygen in a cell culture medium [37], crystallization process [39], mixing of propellants fuels
[35,41], explosives [42-43], milling [44-45], and coating [46-47]. One reported advantage of using the RAM compared to a traditional mixer is a reduction in the mixing time [35, 48-49]. Similarly, it is argued that the RAM, being a self-contained and bladeless device, decreases the shear stress directly applied to the mix, reducing the likelihood of damage to the (AP) particles used in the composite propellant mixture [34, 41]. Finally, since the mixture is self-contained it offers the promise of less waste production, simplified cleaning, and ultimately a cheaper mixing process [50]. The advantages and the drawbacks of the RAM mixer are covered in detail in the review by Wright et al. [35]. Despite the purported benefits of RAM, the mixing performance is not well documented in high solid loading slurry with high viscosity, which is the case for mixing composite solid propellants.

Concrete mixing (high solid loading slurry) using the Ram was evaluated by Vandenberg and Willie [51]. They describe the mixing as occurring in five stages: in the first stage, the motion is dominated by frictional forces between the solids. In the second stage, a wet granular structure is dominated by friction and cohesion forces as the water is wetting the solid and forming liquid bridges. Cohesion forces dominate the (third) following stage, and a hard paste is formed. In the fourth stage, a soft granular suspension is formed and is dominated by cohesive and viscous forces. The microstructure becomes a fluid suspension dominated by viscous forces in the last (fifth) stage. In their work, the mixing process takes place at constant intensity (power), and the acceleration varies according to the mixing stage. Therefore, each stage has a characteristic acceleration response that can be determined with an acceleration vs. time plot. Claydon et al. [42] investigate the mixing performance of the RAM by mixing a plasticizer and a binder with glass microbeads at 62%v/v. The mixing efficiency was measured considering how much time and energy was required to achieve homogeneity. The homogeneity state was defined as the fifth stage described
above, in which a fluid suspension is obtained, and the intensity of mixing (defined as the input power from the mixer) remains constant. The effect of acceleration was determined, and in general, more mixing time and energy are needed to achieve homogeneity at low acceleration.

Zebregs et al. [41] investigated the RAM mixing of HTPB and a bimodal ammonium nitrate (AN) mixture with a solid loading of 81% w/w. Scanning electron microscopy (SEM), density, and ballistic properties measurements were used to compare the RAM and cast-cured process homogeneity. The SEM, density, and burn property results show almost identical outcomes for both methods and, presumably, result in similar homogeneity. The RAM mixing time was reported to be a fraction of the mixing time needed for the cast-cure process. In this work, the size of the particles and the concentration of each type of particle were not specified. The solid loading effect on RAM mixing of aluminum-bismuth(III) oxide in N-dimethyl-formamide (DMF) was investigated by Nellums et al. [43]. A high solid loading produces a minor variation in ignition delays and smaller aggregates compared to low solid loading. Therefore, a high solid loading produces improved performance.

Most of the research done in mixing of slurries using the RAM mixer has been on fluid and solids comprised of only one type of particle. Some of these studies investigate different types of particles blended with a fluid in which the determination of mixing performance was based on characterization by rheology (tensile strength, viscosity) and SEM. In other words, the mixing performance of these slurries is often measured as bulk property (except in the cases where SEM analysis is also performed). A more direct way to measure the homogeneity of the sample, particularly for blends that include a variety of different types/sizes of particles, is through direct determination/measurement of the concentration of each solid component in different regions of
the mixing vessel. To our knowledge, no prior study has comprehensively evaluated the effect of particles sizes and solid composition of the mixing of a slurry using RAM.

Figure 6. RAM velocity field and top view of the mixing vessel and the samples’ location.
4.1 Mixer Performance Determined Experimentally

In this study, we examine the mixing performance of a bimodal mixture of particles within a highly loaded slurry with a high viscosity continuous fluid phase. Specifically, we measure the effect of various size and composition ratios of the particles in the bimodal mixture, the viscosity of the fluid, and two RAM mixing parameters (mixing time and acceleration). In each case, the performance of the resonant acoustic mixer was evaluated by measuring the uniformity of the resulting blend. The uniformity of the blend was determined by comparing the mass fraction and the relative standard deviation (RSD) of the mean mass fraction obtained when a number of samples were systematically analyzed from specific locations within the vessel (see Figure 6). The solid bimodal mixtures used in this study are glass beads of different sizes (ranging from 500 microns to 50 microns); this range was selected because it is commonly used in the preparation of composite propellants fuels [34, 52-53]. The solid loading used is 67% per volume or 81 to 83 % per weight depending on the bimodal mixture and the glycerol solution used, and is chosen to be similar to the weight percent used in a bimodal mixtures employed in practical propellant applications [52]. The liquid used as a substitute for the binder is an aqueous solution of varying amounts of glycerol to control the fluid's viscosity. The results obtained in these experiments can be used to evaluate the mixing performance of the RAM mixer in a high solids loading slurry at different mixing parameters, solid formulation, and viscosity.

4.1.1 Materials and Experimental Set Up

All mixing experiments were performed in a resonant acoustic mixer (RAM) at a selected constant acceleration (g’s). The mixer self-adjusts the resonant frequency, in the range of 58 to 62
Hz, and directly sets the value according to the blend properties. The experiments include high solid loading (67% per volume) of bimodal mixtures in continuous fluids of varying (but high) viscosity. In all the experiments, we use a bimodal mixture of glass particles of different sizes (50, 150, and 500 μm, Polysciences, Inc.) with a solid mass fraction of the tracer particles of 0.20 (not considering the liquid). The variables that were considered include the size of the particles, acceleration (40, 70, and 100 g’s), viscosity (at three values of 9.3751e-7, 3.8447e-5, and 3.8793e-4 m^2/s, respectively) [54], and mixing time from 30 sec. to 8 min.

The tracer particles (0.2 of the mass fraction) were added to the vertical middle of the solid bed. The glycerol solution is added at the top of the solid bed. For the evaluation of the uniformity of the mixture, nine samples were removed from the mixing vessel of 140 ml. The samples were extracted using a theft sampler which extracts samples of approximately 0.75 grams. The locations of the extraction of samples were distributed uniformly around the wall and center of the mixing vessel (as shown in Figure 6). All the samples were removed after the completion of the mixing process at a height equivalent to the vertical middle of the solid bed (i.e., the height at which the tracer particles were initially located). That is, when measuring a time sequence of mixing, we perform a series of identical mixing experiments of increasing duration in order to mitigate the influence of sampling. Each sample was transferred to a glass scintillation vial of 20 mL and dried in a vacuum oven carefully prior to sieving for direct assessment of mass composition.

4.1.1.1 Mass Fraction and Mixing Performance

After the samples were well dried, each sample was sieved with the respective sieve mesh (No.40, 140, and 400), and the weight of both sizes of particles was measured. The mean mass concentration presented in the results is the mean of the measured mass fraction of the tracer particles obtained from the 9 sampling locations.
In addition to recording the mean sample concentration, in order to measure the mixing performance we use also assess the relative standard deviation (RSD) of the mass fraction to measure the uniformity of the blend. The RSD is given by Eq. 4.1:

\[
s = \sqrt{\frac{1}{N-1} \sum_{i=1}^{N} (x_i - \bar{x})^2}
\]

(4.1)

\[
RSD = \frac{s}{\bar{x}}
\]

(4.2)

where \(s\) is the standard deviation of the mass fraction, \(RSD\) is the relative standard deviation, \(x_i\) is the measured mass fractions in each of the 9 samples, \(\bar{x}\) is the theoretical mean mass fraction (i.e., 0.20 in the reported experiments), and \(N\) is the number of samples. In our set of experiments, we use an overall mixture mass fraction of 0.2, and we take nine samples per run in all trials, as stated before.

4.1.2 Results and Discussion

4.1.2.1 Blend Uniformity as Function of Viscosity and Time

In the first set of experiments, we consider the effect of the viscosity on the mixing evolution using a bimodal mixture of 150 μm particles with tracer particles of 500 μm. The viscosity was changed using glycerine-water solutions ranging from pure water (9.3751e-7 m^2/s) and up to 95% per volume of glycerine (3.8793e-4 m^2/s). Figure 7 show the viscosity and mixing time effect in the mean mass fraction and the RSD, respectively. A uniform mixture will exhibit a mean mass fraction of all of the measured samples of 0.2 of the tracer particles and an RSD small or close to 0. The mass fraction measured in the middle of the solid bed will indicate if the
concentration of the tracer particles is well distributed vertically or if vertical segregation is taking place. A high concentration will mean that the tracer particle remains in the initial position. A low concentration will mean that the particles are concentrated in the bottom or the top of the mixing vessel. \( RSD \) is a variability measurement between samples where a low \( RSD \) value indicates that the variation between samples is low.

In general, after two minutes, the RAM shows good mixing performance for the case of lower viscosity (water) even at 40 g’s, in which the mean mass fraction is near the theoretical value of 0.2, and the \( RSD \) is 0.15 or less. For the cases of 75\% and 95\% of glycerol, a higher acceleration is needed to move the solid bed from the bottom of the mixing vessel and have some mixing. For the case of 75\%, the mixing performance improves at 70 g’s in which the \( RSD \) has a minimum value of 0.165, and at 100 g’s is well mixed or uniform with an \( RSD \) of 0.06 and a mean mass fraction of 0.195 after 8 minutes. The case of 95\% at 70 g’s shows that the mixing is poor, with a minimum \( RSD \) of 0.3 and a mean mass fraction of 0.17. In the case of 100 g’s, the mixing performance improves compared to 70 g’s with an \( RSD \) of 0.16 and a mean mass fraction of 0.175. In comparison, the cases of 0\% and 75\% are always more uniform than the 95\% case and can achieve a very uniform blend under certain conditions, and this was not possible at 95\%. The high viscosity mixtures need a higher acceleration and more mixing time than a low viscosity mixture. After 8 minutes, the mixing performance seems to decrease as the \( RSD \) is seen to increase after 12 minutes in multiples cases.
Figure 7. Mass fraction (right) and RSD (left) as a function of time for a bimodal mixture of 150 μm particles with 500 μm particles at A-B)40 g’s, C-D)70 g’s, and E-F)100 g’s.
4.1.2.2 Temperature Effect at High Viscosity

When the fluid is more viscous, the acceleration and mixing time are essential, and this is because the temperature increase in the mixing vessel is dependent on acceleration and mixing time. At high values of the acceleration and mixing time, the temperature increase is significant with changes from 23 °C to 79 °C. For the fluids used in this study, this increase in temperature causes a decrease in dynamic viscosity. As the viscosity decreases, the flow inside the mixing vessel changes from being big granules to a more fluidized solid bed that starts moving as part of the liquid. This is similar to the fifth stage of mixture evolution reported by Vandenberg and Willie [51].

To mitigate or eliminate the temperature effect in the mixing performance, we made two runs at 75% and 95% of a glycerol solution per volume. After 1 minute of operation, we stopped the mixer and let the mixture cool down to room temperature (23 °C) before restarting the mixing process. In this way, the maximum temperature reached was 29 °C during a one-minute session of continuous mixing. As a comparison, in a run of 8 minutes without stopping the mixer, the temperature increased to 79 °C. The acceleration that we used in these cases was 100 g’s because, at this acceleration, the increase in temperature is higher and better results were obtained in the first set of experiments in comparison with 70 and 40 g’s (for the high viscosity systems). Figure 8 compares the results obtained when the mixer ran continuously for 8 minutes -- labelled as T -- and when the mixer was stopped every minute -- labelled as TC. For 75% of glycerol per volume, the mass fraction is found to be near 0.2 of mass fraction in both cases (T and TC), but the RSD for the temperature-controlled case is more than three times the RSD obtained when the temperature was allowed to increase (0.22 vs. 0.066). The increase in RSD means that the variation in the samples is 3 times higher when the temperature is controlled. Therefore, the mixing
performance is worse if the temperature is controlled. In the case of 95% of glycerine, the mass fraction was found to be decreased when comparing the uncontrolled trial to that when the temperature was controlled (from 0.175 to 0.15), and the $RSD$ for the TC and T cases was found to be 0.32 and 0.17, respectively. In both cases, the temperature increase leads to improved mixing performance making the blend more uniform, and the motion of the slurry is visibly observed to exhibit something close to the motion of a fluidized bed. Therefore, the improvement in the mixing performance appears to be caused by the fact that the active viscosity of the blend decreases with an increase in temperature. A better way to cool down the mixing vessel is needed to completely mitigate the temperature effect, like a vessel heat exchanger. Based on these results, the mixing performance is expected to worsen if the temperature is constant and the viscosity is high.
Figure 8. A) Mass fraction and B) RSD with controlled temperature and uncontrolled temperature.
4.1.2.3 Particles Size Effect in Mixing Performance

In order to determine the effect of varying the particle size on the mixing performance, four different combinations of a bimodal mixture of particles were used in water for 8 minutes at 100 g’s. These combinations of particles are 150 μm with 500 μm tracers, 500 μm with 150 μm tracers, 50 μm with 150 μm tracers, and 150 μm with 50 μm tracers particles. Interestingly, the composition effect seems to be very important. If the small particles comprise the minority component (i.e., have the lower mass fraction), as is the case with 500 μm particles with 0.2 of 150 μm tracers, the mixing is quite poor, as seen in Figure 9 (with a measured mean mass fraction of 0.05 and an RSD is 0.81). This is an example of downward segregation, and the small particles are concentrated in the bottom part of the mixing vessel. Another compelling case in which the small particles are the tracer particles is the case in which 150 μm particles with 0.2 of 50 μm tracers particles. In this case, the mixing performance improves when the acceleration is 70 g’s, but the mixing performance gets worse at 100 g’s. We hypothesize that this decrease in performance is caused by the fact that the cohesion forces are more relevant at this size of particles and can enhance the mixing performance [55] provided that the agitation does not mitigate these forces. Thus, at 40 and 70 g’s, the cohesion forces help to mitigate the segregation toward the bottom with an RSD of 0.14 and 0.11. In the case of 100 g’s, the acceleration forces overcome the effect of cohesion forces, and the blend is less uniform with an RSD of 0.34.

In contrast, when the smaller particles are in higher concentration (such as in the blend that contains 50 μm particles with 150 μm tracers or the 150 μm with 500 μm tracers), the resulting mixture is well mixed and uniform under most conditions. The exception is 50 μm with 150 μm tracers at 70 g’s, in which the RSD is 0.32; this means that it is not well mixed. The case of 500
μm with 150 μm tracers was impossible to mix under the conditions investigated. Therefore, we eliminate this combination in our last set of experiments.
Figure 9. A) Mass fraction and B) RSD as a function of the size of the particles in water.
4.1.2.4 Mixing Performance as a Function of Viscosity

The effect of viscosity in a bimodal mixture was investigated at 40, 70, and 100 g’s after 8 minutes of continuous mixing time, and the results are shown in Figure 10. The best mixing performance was obtained with 150 µm with tracers of 500 µm particles. In the case of fluid that include 0% and 75% volume fraction of glycerol, the system achieved a very uniform blend at 100 g’s ($RSD =0.06$ and m.f.$=0.20$), and at this same acceleration an acceptable mixing performance was observed for the fluid containing 95% glycerol (m.f.$=0.175$ and $RSD =0.17$). For the case of 50 µm with a tracer of 150 µm, the viscosity increases the variability in the blend, and the $RSD$ increases. Interestingly, the 95% case is more uniform ($RSD =0.21$ at 100 g’s) compared with the 75% ($RSD =0.49$); we expect that this is caused because, at 75%, part of the solid bed gets stuck to the lid of the container, and the concentration of the big particles is greater there. Therefore, the mass fraction of the tracer particles increases in the solid bed in which the samples were extracted. In the last case of 150 µm particles with 50 µm tracer particles, the viscosity has a big impact at 40 and 70 g’s. Where the mean mass fraction is close to 0.10 at 75% and 95% of glycerine in comparison to the 0% case which is observed to yield a mass fraction of approximately 0.18. At 100 g’s, the mean mass fraction and $RSD$ results are almost the same for all three of the different viscosities. Therefore, the acceleration in the cases with high viscosity improves the mixing performance in general. Interestingly, for the case of water, the $RSD$ increases at 100 g’s, which means that the blend is less uniform than 40 g’s and 70 g’s blends.
Figure 10. Mass fraction (right) and $RSD$ (left) results at different viscosity with binary mixtures of particles with A-B) 150 with 500, C-D) 50 with 150, and E-F) 150 with 50 $\mu$m.
4.1.2.5 Discussion

The impact of uncontrolled temperature (increase) on the mixing performance can be understood by considering the role of temperature on the effective viscosity of the continuous fluid. That is, since the viscosity substantially influences how vigorous is the observed motion within the device, coupled to the fact that the viscosity is a function of temperature, suggests that uncontrolled temperature increase dramatically influences the mixing behaviour. Specifically, when a high viscosity fluid is used in a high solids loading slurry at a controlled (room) temperature, the motion in the mixing vessel will be dominated by granules impact and effective cohesion forces between the grains. In contrast, when the temperature is allowed to increase, the motion will begin to resemble a fluidized bed because of the concomitant decrease in viscosity. Therefore, it is hard to break up the solid granules with a low temperature, and the mixing performance is lower, while a higher temperature improves the mixing outcome. As seen in the work of Claydon et al. [42], much of the mixing occurs when a “churning stage” is achieved where a bulk rolling motion produces bulk mixing. If the temperature is controlled at room temperature, this stage is not achieved (with a high viscosity fluid), and the resulting mixing performance is bad. At high temperatures, a fluidized solid bed (“churning stage”) increases the particles’ motion and improves the mixing performance.

Segregation (or unmix) occurs when a difference in mechanical material properties between particles is present (size difference in our case). Size segregation is observed in different types of mixers and has not previously been evaluated in the RAM mixer. In this study, we note differing segregation behaviour as a function of both fluid viscosity as well as relative composition of small/large particles. For example, size segregation toward the bottom was observed at some viscosities when the small particles were the tracer particles. In a true granular flow, this type of
segregation is known as percolation [30], where the small particles move through the void spaces between big particles. As a result, at the end of the mixing process, the small particles are mainly in the bottom of the mixing vessel. Percolation was observed for the case of 500 μm and 150 μm tracer particles in low viscosity (water) where the mean mass fractions measured in the middle of the mixing vessel dropped to 0.05, far from the actual mass fraction of 0.2. Also, percolation was observed in the case of high viscosity for a mixture comprised of 150 μm with 50 μm tracer particles at low acceleration (40 and 70 g’s) with an RSD >0.4 and a mean mass fraction close to 0.10. For this same system, at a high acceleration, the mixing performance was not viscosity dependent and the segregation decreases at high viscosity.

Another interesting result was that in cases where we used 75% glycerol within the solution, the mixing performance was worse than what is observed in the case of a 95% glycerol solution with particles that included 50 μm bulk with (larger) tracers of 150 μm. When using a 75% glycerol solution, we reported that part of the solid bed was stuck to the lid of the vessel because the motion inside the mixing vessel was turbulent, and “splashing” was observed [57]. This motion regime is not optimal for mixing high solids loading slurries. For the 95% case, the motion regime that dominates was a viscous suspension motion without splashing; therefore, a higher mixing performance is observed than in the 75% case.
4.2 Modeling of RAM with our LBM-DEM Model

4.2.1 Macromixing Results

A bimodal mixing simulation was done using two combinations of 1000 particles with a diameter of 2.4 mm and 0.8 mm (same ratio size as the case of 500 μm with 150 μm used in the experiments) with 24% of the tracer particles. Tracer particles were located in the center of the solid bed, similar to the experiments. The simulation grid used was $N_x=100$, $N_y=150$ and $N_z=100$ nodes and a relaxation parameter $\tau = 0.75$. The dimensions of the simulated container are the same as the original container used but in a rectangular shape with the following dimensions 38 mm in width, 38 mm in depth, and 57 mm in height. In these simulations, the acceleration used was 10 g’s and 20 g’s, and the viscosity used was $0.00002e^{-5}$ m^2/s (75% glycerin) and $3.8793e^{-4}$ m^2/s (95% glycerin). $RSD$ was measured for all the particles in an area of five-time the radius of the small particles. In this area, each type of particle was measured, and this concentration inside the area was used to determine the $RSD$.

The effect of having the small particles as a tracer in the mixing performance is very noticeable in the simulations. The $RSD$ is more than two times higher when the bigger particles are the tracer (Figure 11 A) and B)). These results match the trend observed in the experimental case with 500 μm with 150 μm particles as a tracer, which was segregated toward the bottom, and the mixing performance was poor. The acceleration and viscosity did not significantly impact the case of 0.8 mm with 2.2 mm particles since the $RSD$ is almost the same for all the cases ($RSD$ between 0.14 to 0.16). In contrast, for the case of 2.2 mm with 0.8 mm particles, the trends in the results are similar to the trend observed experimentally because the acceleration makes the mixing blend more uniform, decreasing the $RSD$. In this case, increasing viscosity worsens the mixing.
performance, increasing the $RSD$ (less mixed). This trend was the general pattern observed experimentally.

Therefore, our model can capture the effect of acceleration, bimodal distribution size, and viscosity. This model has three limitations: the change in viscosity due to the increases in temperature caused by the acceleration and particle collisions, high acceleration simulation due to restriction of the lattice Boltzmann method, and the number of particles.

The experimental value of the $RSD$ and the value obtained from the simulation cannot be compared directly for multiple reasons: the size of the particles is not the same, the acceleration is not the same, and the method to calculate the $RSD$ is different. The full mixing vessel was analyzed in the simulation compared with less than 10% of the particles in a specific region in the experiment. Yet the trends in the model results are similar to the trend of the results obtained experimentally.

Figure 11. Change in RSD with respect to time for A) 0.8 mm with 2.4 mm as a tracer and B) 2.4 mm with 0.8 mm as a tracer.
4.3 Conclusion

In this chapter, we evaluated the mixing performance of the RAM by considering the effect of the mixing time, viscosity, particle sizes, solid composition, and acceleration. Mixing time was important when the viscosity was high, and in general, more mixing time is needed to have a good mixing performance than when running low viscosity mixtures, which require less mixing time. In general, better mixing performance was observed when using fluids of lower viscosity. The particles size (and relative composition) was also found to be essential to the mixing performance, and segregation toward the bottom of the mixing vessel was observed when the small particles were in a lower concentration. Also, one specific case with 500 μm particles with 150 μm tracer particles was impossible to mix under the investigated conditions. In general, the mixing performance was better when the big particles had a lower concentration than the smaller particles (i.e., in these cases a more uniform mixture was obtained). High acceleration is essential to obtain a good mixing performance when employing a high viscosity fluid. The main reason is that a high acceleration increases the mixing vessel temperature and this increased temperature results in a decrease in the dynamic viscosity. As a consequence, the observed mixing improves, and the slurry is fluidized. Future work in exploring the RAM mixing performance should include examination of the potential for density segregation, where heavy particles are usually concentrated at the bottom.
5.0 Low Viscosity Liquid Bridges: Stretching of Liquid Bridges Immersed in a Higher Viscosity Liquid

A liquid that preferentially wets particles can form a meniscus that bridges across two particles. Such a liquid bridge often induces attraction between the particles, and this attraction is defined as attractive capillary forces. Capillary forces are relevant across multiple processes: material processing [58], granular materials [59], soil mechanics [60], and concrete mechanics [61]. We now have an excellent quantitative understanding of capillary forces in a liquid bridge under static conditions [62-63]. While most of the past literature has been for the case when the particles are preferentially wetted by the particles [64-66], the case of less-wetted particles (which can induce repulsive forces) has also been examined [67-68]. A numerical method was used by Lian and Seville [63] to develop an equation that predicts the capillary force for a given liquid bridge volume and finite solid-liquid contact angle.

Describing the force in most of the processing operations as a static meniscus force is inadequate because this process involves relative motion between particles. With motion, the viscous force needs to be taken into consideration. For example, in a fluid bed granulator (FBG) mixer, a liquid binder is applied to a powder mix to form granules under a shear flow, and the viscous force plays an important role. The first step of the granulation is when the binder forms liquid bridges between particles and binds them together by a combination of capillary and viscous forces [59]. If the relative velocity between the particles is large, the viscous forces may far exceed capillary forces, which is common in multiple industrial processes. Matthewson [69] derived an equation for the viscous force component in a liquid bridge between a particle and a flat plate. The viscous component was investigated as an impulse required to separate the two surfaces and was
combined with the meniscus force to determine the total force. Ennis [70] et al. investigate the effect of the capillary number (Ca) on the strength of the liquid bridge between two spheres. They reported that when Ca<0.0035, the viscosity effect can be neglected, and when the Ca> 0.35, the viscosity effect is important. Pitois et al. [71] experimentally measured the capillary and viscous force as a function of the separation velocity of two spheres. They found that the rupture distance increases with separation velocity. Under dynamic separation, viscous force and velocity are essential in the liquid bridge rupture force and distance [71-72].

All of the mentioned studies were done with air as a continuum fluid. Mason and Clark [64] investigated the strength of an oil liquid bridge between two particles in water as the surrounding fluid, considering the volume of the liquid bridge and the separation distance. A maximum in force was reported at a short separation distance. These maximum in force were in good agreement with the theoretical value, and an exception was reported when the liquid bridge had a smaller volume. Rosetti et al. [73] investigate the rupture energy of silicon oil liquid bridge formed in water between two spheres under quasi-static conditions (i.e., low Ca). They calculated the energy at rupture distance and a short separation distance. In this study, particles with high wettability with the liquid binder show a liquid bridge with higher energy. The capillary number only affects the force calculation when Ca> 10^-4. Uguz et al. [74] use experiments to study the stability of a liquid bridge between two plates surrounded by a solution of glycerol in a shear flow. Results show that the stabilization using shear flow is possible and that the liquid bridge is stable past its static critical point.

In the above literature review, most of the model and experimental studies of the liquid bridges were done with a viscous liquid bridge, usually in the air or in a less viscous continuum fluid. Research about the breaking bridge of a less viscous liquid inside a more viscous liquid is
sparse, and this type of liquid bridge can be seen during material processing. Spherical crystallization has been applied in different industrial sectors, in the pharmaceutical industry to make crystals used to make tablets [75], in the food industry to remove specific particles [76], and in the processing of wastewater to remove heavy metals [77]. In spherical crystallization, a binder, sometimes less viscous than a continuum phase, is added to create liquid bridges between particles and produce particles agglomerations or crystals depending on the application [78-79]. Similarly, capillary forces can be used in particle suspensions to induce aggregation of particles in capillary suspensions. For example, mixing a small amount of water into a suspension of hydrophilic particles in oil can induce the aggregation of particles via liquid bridges of water. The resulting "pendular state" suspension can have a yield stress [80-81]. Such suspensions are considered precursors to porous ceramics [82], especially for 3D printing [83]. Similar capillarity-driven aggregation can be induced in particle-filled polymer blends, e.g., to realize conductive plastics or adhesives [84-85]. In such cases, the fluid added to the particles may be much less viscous than the continuous phase polymer, and there is little knowledge of the behavior of such low-viscosity bridges.

A surrounding fluid more viscous than the bridge affects the dynamic of liquid bridge rupture because as the bridge is elongated, the more viscous fluid will fill the narrow gap between the sphere and the plate. This paper is a modeling and experimental study of forces associated with the stretching and rupture of low viscosity liquid bridges surrounded by a higher viscosity external fluid. We investigated the effect of a viscosity match meniscus in the total force for comparison purposes. In next section, we propose a model for the force for normal motion between a particle and a flat plate. Section 3 describes the experimental, and Section 4 the results for measured forces during bridge stretching. Section 5 concludes with a brief discussion and summary.
5.1 Theory

5.1.1 Viscous Force

Consider the geometry (Fig. 12) of a plate and a sphere of radius \( R \), at a separation \( S_0 \) immersed in an outer fluid of viscosity \( \eta_o \). Considering first the case without a liquid bridge, the lubrication equation using the well known Reynolds’ approach for the pressure for normal-direction motion in an axisymmetric geometry is given by [69]

\[
\frac{d}{dr} \left( rS^3(r) \frac{dP}{dr} \right) = 12\eta_orv \tag{5.1}
\]

where \( v \) is the separation velocity and \( \eta_o \). At close separation \( (S_0 \ll R) \), the geometry can be approximated by \( S(r) \approx \frac{r^2}{2R} + S_0 \). One can then integrate Eq. 5.1, and set \( P = 0 \) far away (i.e. at \( r \to \infty \)), and \( dP/dr = 0 \) at \( r = 0 \) to yield the pressure profile \( P(r) \)

\[
S(r) = -12 \frac{\eta_o R^3v}{(2RS_0 + r^2)^2} \tag{5.2}
\]

Integration of the above pressure profile yields the viscous force [86]

\[
F_{vis} = -6\pi \eta_o R^2 \frac{1}{S_0}v \tag{5.3}
\]
Note that for positive velocity (i.e. stretching the liquid bridge), the pressure and force are both negative indicating that the force is attractive. Turning now to the geometry of Fig. 1B, consider a liquid bridge of viscosity $\eta_i = \eta^* \eta_o$ of volume $V$ occupying the region $r < b$. Here $\eta^*$, dubbed relative viscosity, is the ratio of the viscosity of the inner to the outer fluid. For the purposes of calculating the liquid volume, we assume that the bridge has a cylindrical edge, and write

$$V = \int_{0}^{b} 2\pi r S(r) dr \equiv \pi R \left[ S^2(b) - S_0^2 \right]$$ (5.4)

Here above approximation $S(r) \approx \frac{r^2}{2R} + S_0$ was used. The volume can be rendered non-dimensional as

$$V^* = \frac{V}{\pi RS_0^2} = \frac{S^2(b)}{S_0^2} - 1$$ (5.5)

Accordingly, $S(b)$ can be written in terms of volume as

$$S(b) = S_0 \sqrt{V^* + 1}$$ (5.6)

We will now derive the pressure profile for the case when $\eta_i \neq \eta_o$. For the outer fluid, the pressure profile is still given by Eq. 5.2:

$$P_o(r) = -12 \frac{\eta_o R^3 v}{(2RS_0 + r^2)^2} \text{ for } r > b$$ (5.7)

where $P_o$ is the pressure in the outer fluid. By substituting $r = b$, we can obtain the pressure at the edge of the bridge:

$$P_o(b) = -12 \frac{\eta_o R^3 v}{(2RS_0 + b^2)^2}$$ (5.8)
For the inner fluid, Eq. 5.1 is still valid, but simply replacing \( \eta_o \) by \( \eta_i \) and integrated, but now setting the boundary condition at the edge of the bridge as per Eq. 5.8 to impose continuity of pressure

\[
P_i(r) = -12 \frac{\eta_i R^3 v}{(2RS_0 + r^2)^2} + 12 \frac{(\eta_i - \eta_o)R^3 v}{(2RS_0 + b^2)^2}
\]

for \( r < b \)  

(Eq. 5.9)

Eqs. 5.7 and 5.9 together give the pressure profile in the entire domain. Now the total viscous force can be obtained by integrating the overall pressure profile:

\[
F_{vis} = \int_0^\infty 2\pi \Delta P r dr = \int_0^b 2\pi \Delta P i r dr + \int_b^\infty 2\pi \Delta P o r dr
\]

(Eq. 5.10)

The final expression is:

\[
F_{vis} = -\frac{6\pi \eta_0 R^2 v}{S_0} \left( \frac{V}{\sqrt{\pi R}} + S_0^2 S_0 \right) \left( \eta_i \frac{V}{\sqrt{\pi R}} + S_0^2 S_0 (\eta_o - \eta_i) \right)
\]

(Eq. 5.11)

Here the first and the second terms on the right-hand side are the viscous contributions of the outside and bridge fluids respectively. This final expression can be written out in non-dimensional terms as

\[
F_{vis} = -\frac{6\pi \eta_0 R^2 v}{S_0} \left( -\frac{1}{\sqrt{V^* + 1}} + \frac{(\sqrt{V^* + 1} - 1)(\eta^* \sqrt{V^* + 1} + 1 - \eta^*)}{V^* + 1} \right)
\]

(Eq. 5.12)

where

\[
\eta^* = \frac{\eta_i}{\eta_o}
\]

(Eq. 5.13)
The quantity outside the square brackets in Eq. 5.12 is the viscous force from Eq. 5.3, whereas the term in the square brackets factor that accounts for the geometry and viscosity of the liquid bridge, captured by the non-dimensional variables $V^*$ and $\eta^*$ respectively. This multiplicative factor is less than 1 when $\eta^* < 1$ and greater than 1 when $\eta^* > 1$. It must be emphasized that only quantities that are directly controlled experimentally appear in Eqs. 5.11 and 5.12; $b$ and $S(b)$ do not appear.

Eq. 5.12 recovers Eq. 5.3 in the limit of $V = 0$ or $\eta^* = 1$. It also recovers the equation derived by Pitois for $\eta^* \gg 1$. It is crucial to recognize that the region covered by the bridge still contributes to the viscous force even if the bridge fluid is inviscid. In this limit, the pressure inside the bridge is simply $P(b)$, and hence the bridge contribution to the viscous force is $\pi b^2 P(b)$. This can be verified readily by setting $\eta^* = 0$ in the latter term in Eq. 5.12.

### 5.1.2 Capillary Force

The static capillary force for of small liquid bridge with fixed volume in between a sphere and a plane (Figure 12) is given by [65]

$$F_{cap} = -4\pi R \gamma \cos \theta \left( 1 - \frac{1}{\sqrt{V^* + 1}} \right) \quad (5.14)$$

where $\gamma$ is the interfacial tension, and $\theta$ is the contact angle. This equation is valid when the top solid/liquid bridge contact angle is small, and therefore, the axial component of surface tension can be neglected. Also, it is only valid when the separation distance is smaller than the rupture distance of the liquid bridge ($S_0 < S_{rup}^0$).
5.1.3 Total Force

Following Pitois, we assume that the total force can be obtained from the sum of the static capillary force (Eq. 5.14) and the viscous force (Eq. 5.12):

\[ F_{tot} = F_{cap} + F_{vis} \] (5.15)
Figure 12. Schematic of experimental apparatus measuring the force during stretching of a liquid bridge between a sphere and a plane. Note that there is no macroscopic motion of the fluid. B. Schematic defining the various geometric quantities of the bridge.
5.2 Experimental Procedure and Materials

We seek to quantify viscous effects when the viscosity of the liquid bridge is lower than of the continuous phase. Thus, not only is the bridge submerged, but the submerging liquid has a relatively high viscosity. This requires the force-sensing mechanism (a cantilever in our case) to be mounted outside this high viscosity liquid; otherwise, viscous drag on the mechanism would affect the measurements.

5.2.1 Experimental Apparatus

Figure 12 shows a schematic of the experimental apparatus where the particle is attached by a long pin of diameter 0.8 mm to a cantilever. The deflection of the cantilever can be measured to sub-micron accuracy using an optical displacement sensor (Philtec) which measures light reflected from the back of the cantilever. The apparatus is mounted in an active air vibration insolation table (Kinetic Systems). Below the particle is mounted a planar surface whose vertical position is controlled with a stepper motor (Moons Industries). It is crucial to recognize that the particle-plane separation is changed without translating the fluid bath, thus there is a negligible change in height at the upper surface of the liquid. This is essential to the current experiment; any gross motion of the air-liquid interface induces capillary and gravity forces on the surface of the pin holding the particle, which overwhelm the forces of interest.
5.2.2 Materials

Polyisobutylene (PIB 24, Soltex) was used as the outer continuous phase fluid in all experiments. Most experiments used polyethylene glycol (PEG, molecular weight 600 g/mol) as the low-viscosity bridge fluid. A limited number of experiments were conducted using polydimethylsiloxane (PDMS, Rhodia), whose viscosity is close to that of the PIB. The properties of each fluid used are available in Table 3.

The particles were glass spheres of diameter 4 mm. When using PEG as the bridge fluid, the particles were preferentially wetted by PEG. When using PDMS as the bridge fluid, the particles were coated with a thin layer of Sylgard 184 silicone rubber to ensure near-complete wetting by the PDMS fluid.

<table>
<thead>
<tr>
<th>Liquids</th>
<th>Viscosity (Pa s)</th>
<th>Interfacial tension (mN/m)</th>
</tr>
</thead>
<tbody>
<tr>
<td>PIB 24 (surrounding fluid)</td>
<td>30.0</td>
<td>---</td>
</tr>
<tr>
<td>PEG 600</td>
<td>0.156</td>
<td>10.5</td>
</tr>
<tr>
<td>PDMS Oil</td>
<td>35.3</td>
<td>2.5 [87]</td>
</tr>
</tbody>
</table>

5.2.3 Experimental Procedure

A drop of the bridge fluid of the desired volume was first placed on a glass slide in air with a micropipette (for PEG bridges), or with a plastic toothpick (PDMS bridge). In the latter case, the bridge volume was determined using the drop's weight. The slide was placed on the horizontal platform. It was then immersed into the reservoir of PIB, immediately under the glass particle. The particle was brought into contact with the plate and allowed to rest for 4 minutes to let the bridge
equilibrate. The platform was then moved downwards at a separation velocity in the range of $\nu = 1 \, \mu\text{m/s}$ to $320 \, \mu\text{m/s}$. The experiments were video-recorded with a Dino-Lite digital microscope. The corresponding cantilever deflection was converted into force using the cantilever spring constant.

The cantilevers were 3D-printed plates whose thickness was selected based on a tradeoff between the noisiness in the data and the maximum cantilever deflection that can be tolerated. At low speeds or under quasi-static conditions, the forces encountered were low and hence softer cantilevers were needed to achieve an adequate signal-to-noise ratio. At higher speeds, the deflection of these softer cantilevers becomes a significant portion of the applied displacement, which is unacceptable. Thus, higher velocities needed stiffer cantilevers. The cantilever spring constant was calibrated using deadweights hung from the cantilever at the same position as the pin holding the particle.

In our calculations, the contact angle was assumed to be $0^\circ$ in the theoretical force equation (Eq. 5.14 and 5.15).
5.3 Results

5.3.1 Viscous Force with no Meniscus

The viscous forces during the separation of the sphere from the flat plate in the absence of a liquid bridge are shown in Figure 13 A). As in all the experiments in this work, the continuous phase was PIB 24, and the initial separation was $S_0 = 50 \, \mu m$. As expected, the viscous force reduces during separation, and reduces with decreasing separation velocity, $v$. In the absence of a liquid bridge, we expect the force to be proportional to $v$, and hence Figure 13 B) plots the ratio of force to velocity. As expected, data from different velocities collapse, and are in good agreement with Eq. 5.3 with no fitting parameters. These results prove that our experimental device can precisely capture the force applied to the sphere and the effect of velocity.
Figure 13. A) Viscous force at different velocities between a sphere and a moving flat plate starting at an initiation separation of $S_0 = 50 \mu m$ of separation distance. B) The ratio of force to velocity collapses all the results into a single curve. Dashed line in both graphs is Eq. 5.3.
5.3.2 Meniscus Evolution

Before discussing forces, we show how the meniscus evolves with time and discuss the effect of velocity on the meniscus evolution. A liquid bridge of $V = 5 \, \mu\text{L}$ of PEG 600 was added between the sphere and the glass slide, and the initial separation was set at $S_0 = 50 \, \mu\text{m}$. A first experiment was conducted at a separation velocity $v = 1 \, \mu\text{m/s}$, and the images in Figure 14 A)-D) were recorded. The initial separation was then reset to 50 $\mu$m, and the liquid bridge allowed to equilibrate for 4 minutes before the experiment was repeated at $v = 100 \, \mu\text{m/s}$, and the images in Figure 43 E)-H) were recorded. The recession of the meniscus is clearly different in both cases. At low velocity, the meniscus has more time for equilibration (indeed we will show later that this experiment may be regarded as quasi-static). Accordingly, the meniscus fluid is sucked out of the liquid bridge, both due to capillarity as well as gravity, leading to a thin bridge which ruptures when $S_0$ is roughly 1600 $\mu$m. In contrast, high separation velocity results in a fatter liquid bridge which does not rupture even at $S_0 = 2000 \, \mu\text{m}$. This increase in rupture distance with velocity will be covered in detail in Section 5.3.6.

In all experiments, a portion of the bridge fluid remains as a layer on the bottom of the particle, whereas a larger portion remains coated on the plate, presumably due to gravitational effects. The portion left on the plate decreased with an increase in separation velocity. This pattern has been reported previously for liquid bridges between two flat plates [89], and that gravity has most influence at low velocity. The influence of gravity in this situation is generally captured by the Bond number, which is the ratio of gravitational force over the surface tension force in the liquid bridge:
where $\Delta \rho$ is the density difference, and $g$ is the acceleration constant. The Bond number for the $V$ values in this paper (1 to 5 $\mu$L) is in the range of 0.22 to 0.65. Incidentally it is common to instead define a Bond number based on the dimension $b$. If we define $Bo$ as $\frac{b^2 \Delta \rho g}{\gamma}$, then using the $b$ values calculated from $V$ at a separation distance of 50 $\mu$m, the $Bo$ values are in range of 0.3 to 0.72. With either definition, $Bo$ is on the order of 1 suggesting a moderate effect of the gravitational forces, at least under static conditions.

Figure 14. Liquid bridge of PEG 600 with a volume $V = 5 \mu$L undergoing separation at velocity $v = 1 \mu$m/s (upper row) and $v = 100 \mu$m/s, (lower row). The $S_0$ values for the first three images in each row are listed at the top. Image D) is at $S_0 = 1557 \mu$m, immediately after rupture. Image H) corresponds to $S_0 = 2000 \mu$m and the bridge has not yet ruptured.
5.3.3 Effect of Meniscus Viscosity on the Force During Separation

The key question underlying this research is about how low viscosity bridges affect the total liquid bridge forces. Figure 15 illustrates the main effects qualitatively. We compare the measured force in three cases: no meniscus, viscosity-matched meniscus (PDMS), and low viscosity meniscus (PEG) where the latter two have roughly comparable volumes. In all cases, the separation velocity was \( v = 100 \ \mu m/s \), and the initial separation was \( S_0 = 50 \ \mu m \). As will be shown later, at this velocity, capillary forces are much smaller than viscous forces, thus allowing the effects of meniscus viscosity to be identified clearly.

The forces during particle separation for the viscosity-matched meniscus are similar to those obtained with no meniscus since the viscosity is the same for the surrounding fluid (PIB) and the meniscus (PDMS). The high viscosity meniscus shows a higher force in comparison to the low viscosity meniscus and the no meniscus case. Since the viscosity of both fluids is the same (PDMS and PIB 24), the total force for this case is equal to the viscous force (Eq. 5.3) plus the capillary force (Eq. 5.14). Comparing the viscous meniscus case with the no meniscus case, since the interfacial tension between PDMS and PIB is 2.5 mN/m, a slightly higher total force was obtained experimentally because of the added capillary force with the meniscus. The measurements are in good agreement with the theoretical force and the total experimental force with viscosity match.

In contrast, the low viscosity bridge shows much lower forces at small separations, which is in qualitative agreement with the theoretical model. To our knowledge, this is this is the first documentation in the literature of a low viscosity liquid bridge lowering the total force during separation. Quantitatively however, eq. 5.15 overestimates the total force for the case of low viscosity meniscus by a factor of \( \sim x2 \), this will be discussed further in the discussion section.
Apart from the forces, there is also a sharp difference between the evolution of the liquid bridge profile where the low viscosity bridge rapidly thins to a much smaller diameter until at a $S_0/R$ value of 0.7, the bridge comprises a slim filament of fluid, whereas the high viscosity meniscus is far wider. It appears therefore that the low viscosity meniscus leaves behind a relatively thick coating of liquid on the particle and on the bottom plate.

The rupture distance was not measured for the viscosity match meniscus but is higher than the rupture distance of the low viscosity meniscus. This longer rupture distance is expected because the rupture distance depends on the liquid bridge viscosity.
Figure 15. A)-D) and E)-H) images of the PEG and PDMS meniscus respectively at different $S_0/R$ (labeled at the top of each column the second row). 1)Total force for a meniscus of 2.68 $\mu$L of PDMS, 2 $\mu$L of PEG, and no meniscus at a separation velocity of 100 $\mu$m/s, the respective theoretical value using Eq. 15. The dark black region in A-D is an ink-mark made on the front edge of the bottom plate and does not interfere with this experiment. It was placed during early experiments to track motion of the bottom plate.
5.3.4 Effect of Velocity

Figure 16 A) shows the effect of separation velocity on the force evolution for the low viscosity PEG bridge at a bridge volume of $V = 2 \mu\text{L}$. The force increases with increasing velocity indicating increasing viscous contributions. In contrast, the capillary contributions are expected to depend primarily on separation; in fact, the model assumes that the capillary forces are equal to the static capillary force, and therefore are velocity-independent. Therefore, two regimes may be distinguished: a viscosity-dominated regime corresponding to $F_{\text{tot}} \propto v$ and a capillary-dominated regime corresponding to $F_{\text{tot}}$ being independent of $v$. Accordingly, Figure 16 B) plots the ratio of force to velocity and shows that speeds above $v = 60 \mu\text{m/s}$ are viscosity-dominated, whereas below that velocity, surface tension makes a significant contribution to the total force. In the viscosity-dominated regime, the total force $F_{\text{tot}}$ in Eq. 5.15 may be approximated as $F_{\text{vis}}$ since the capillary force is negligible; however, Figure 16 B) shows that the data lie significantly below the model. This is discussed further in Section 5.4
Figure 16. A) Force curves during separation for a liquid bridge of volume $V = 2 \, \mu \text{L}$ at different separation velocities and the predictions of Eq. 15 (dot-dashed lines, where the line color corresponds to each symbol color). B) Ratio of the force divided by velocity compared to the total force Eq. 5.15.
5.3.5 Effect of Meniscus Volume

Finally, we turn to examining the effect of bridge volume on the separation forces. Eq. 5.15 predicts that with increasing bridge volume, the viscous contribution to the separation force decreases. This is because the bridge spans a wider region (i.e. a larger $V$ implies a larger $b$), and hence more of the near-contact region is occupied by the low viscosity fluid. On the other hand, a larger bridge volume raises the rupture distance $S_{0\text{rup}}$, and allows the capillary force to persist to larger values of $S_0$. Thus, experiments must be conducted at both high and low velocities to evaluate the differing effects in the viscosity-dominated vs capillary-dominated regimes.

Figure 17 A) shows that at high velocity, the separation force is much lower than the case without a liquid bridge as shown previously in Figure 15. Further, the degree of decrease in force increases with increasing bridge volume. Beyond a nondimensional separation of about $S_0/R$ of 0.6, all three volumes have comparable forces suggesting that the bridge makes little contribution to the total force at larger separations.

In the opposite extreme, at $v = 5 \ \mu m/s$, the situation is reversed. The viscous forces are relatively small, and for the $V = 5 \ \mu L$ liquid bridge, the separation force exceeds that of the non-meniscus case, and further, the force decreases only gradually as separation increases. This is a consequence of the relatively large bridge volume which allows the bridge to survive without rupturing up to a large distance. Similar results were observed previously for the case of a viscous meniscus with air as the surrounding fluid in which the capillary force dominates the viscous force with an increase in separation distance [66][71]. At lower bridge volume $V = 1 \ \mu L$, the force approaches zero because the bridge ruptures beyond $S_0/R=0.5$, and the viscous forces are small at $v = 5 \ \mu m/s$.  

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The case of $v = 40 \ \mu m/s$ appears intermediate between these two extremes. The liquid bridge lowers the force at small separations when viscous force dominates, but not at large separations. Indeed at large separation, the bridge may increase the force slightly because of capillary contributions.
Figure 17. The total force exerted in a particle with a PEG bridge volume of $V = 1 \ \mu\text{L}, 5 \ \mu\text{L}$ and no meniscus using a separation velocity of A) $v = 320 \ \mu\text{m/s}$, B) $40 \ \mu\text{m/s}$ and C) $5 \ \mu\text{m/s}$). Note that the y-scale changes several fold going from A) to D).
5.3.6 Liquid Bridge Rupture Distance

Finally, our experiments also show the velocity-dependence of the distance $S_{0, rup}$ at which the liquid bridge ruptures. These are shown in Figure 18 A) for bridges of volume of $V = 1$ and 5 µL. The lowest speed shown in Figure 18 A) is $v = 1 \mu m/s$, at which the rupture distances are in reasonable agreement with the values estimated from Eq. 5.15. The maximum velocity shown in Figure 18 A) is $v = 100 \mu m/s$ since beyond this velocity, the liquid bridge draws into a very thin filament prior to breaking, making it difficult to determine the specific instant at which the liquid bridge breaks. At both bridge volumes, the rupture distance increases significantly as velocity increases. Similar results have been seen by Pitois et al for viscous bridges between particles, and by Zhang and Basaran for inertial bridges between parallel plates [89].

Pitois et al. [71] observed that for a liquid bridge of high relative viscosity, the results could be well-captured by

$$ \Delta S_{0, rup} \propto \frac{1}{v^2} \quad \text{(5.17)} $$

where

$$ \Delta S_{0, rup} = \frac{S_{0, rup} - S_{0, stat, rup}}{S_{0, stat, rup}} \quad \text{(5.18)} $$

$$ S_{0, stat, rup} = \left( 1 + \frac{\theta}{4} \right) \left[ \left( \frac{V}{R^3} \right) - \left( \frac{2}{5} \frac{V}{R^3} \right) \right] \quad \text{(5.19)} $$

and $S_{0, stat, rup}$ is the rupture distance at static conditions, i.e. in the limit of $v \to 0$. Eq. 5.19 is only valid for a liquid bridge between sphere-sphere. In non-dimensional terms, Eq. 5.17 suggests that $S_{0, rup} \propto Ca^{0.5}$ where $Ca = vR/\gamma$ is the capillary number based on particle size. To test if our
measurements follow Eq. 5.17, we calculated $\Delta S_0^{rup}$ assuming that the case of $v = 1 \, \mu m/s$ was sufficiently slow that it corresponded to static rupture conditions. The $\Delta S_0^{rup}$ thus-calculated are plotted against $Ca$ in Figure 18 B)), and are in reasonable agreement with the square root dependence Eq. 5.17.

It is critical to point out that regardless of whether Eq. 5.17 is followed or not, there is a large quantitative difference in these results vs those for high viscosity bridges [71], which are added to Figure 18 B) for comparison. In Figure 18, $\Delta S_0^{rup}$ has relatively large magnitudes, approaching 100%. In contrast, the largest value of $\Delta S_0^{rup}$ noted by Pitois was about 10%.
Figure 18. Rupture distance ($S_0^{\text{rup}}$) as a function of separation velocity and and B) normalized rupture distance ($\Delta S_0^{\text{rup}}$) vs capillary number for a PEG liquid bridge, and data from Pitois et al. C-E. Images of liquid bridge immediately prior to rupture at different velocities C) $v = 2 \mu m/s$, D) $20 \mu m/s$ E) $40 \mu m/s$ and F) $80 \mu m/s$. Dashed lines, indicating the edge of the particle and the location of the plate, illustrate the increase in rupture distance with velocity.
5.4 Discussion

To summarize briefly, we consider the elongation of a liquid bridge between a spherical particle and a flat plate. This chapter develops a model for the viscous forces when the bridge viscosity differs from the viscosity of the continuous fluid. The model and experiments both show that the separation forces decrease significantly when the liquid bridge has a much lower viscosity than the surrounding fluid.

This section addresses two issues. The first issue is the distinction between viscous-dominated vs capillary-dominated regimes of separation. In Figure 16 B), this distinction was made empirically based on whether the force is proportional to velocity or not. A theoretical approach however is to take the ratio of the viscous to capillary forces, which based on Eqs. 5.3 and 5.14, result in an equation of the form

\[
\frac{F_{\text{vis}}}{F_{\text{cap}}} = \frac{v \eta_o R}{\gamma S_0} \mathcal{F}(V^*, \eta^*) = C a \frac{R}{S_0} \mathcal{F}(V^*, \eta^*)
\]  \hspace{1cm} (5.20)

The function \( \mathcal{F} \) can be deduced from Eqs. 5.3 and 5.14. In some situations, e.g. coating operations, breakup of drops and bubbles, the viscous-dominated vs capillary-dominated regimes may be identified solely based on whether \( Ca \) is much larger or much less than 1. In the present situation however, it is clear that the ratio of viscous to capillary forces increases as \( S_0 \) reduces. This is true regardless of whether the bridge viscosity is lower or higher than that of the outer fluid. Accordingly, if the particles are initially in contact (which would generally be the case for freely-suspended particles bonded by a liquid bridge), the separation process always starts in a viscosity-
dominated regime no matter how small the velocity. During the separation process, \( \frac{F_{\text{vis}}}{F_{\text{cap}}} \) may become less than 1, i.e. the separation process may transition to a capillary-dominated regime. This point – that the initial separation is always viscosity-dominated – applies regardless of whether \( \eta_o \) is more or less than \( \eta_i \), and does not appear to be appreciated in the previous literature.

The second is the discrepancy between predictions and experiment. One may intuitively expect, and indeed Eqs. 5.12 and 5.13 predict, that a low viscosity bridge would reduce the force required for separation. However the observed effect is even larger than predicted: for low viscosity bridges, the experimentally-measured forces are typically 2x smaller than the predictions. This is true even at small separations when the lubrication approximation, Eq. 5.1, is most likely to be valid. In contrast, Eq. 5.12 is in good agreement with experiments for a viscosity-matched bridge (Figure 15), and/or for a high viscosity bridge [71]. We speculate that the reason for the discrepancy at low \( \eta^* \) is that the lateral extent of the liquid bridge exceeds that assumed in the theory. In deriving Eq. 5.4 (which relates the extent \( b \) of the liquid bridge to its volume), we implicitly assumed that the contact line of the liquid bridge recedes as the separation distance increases (compare Figure 19 A) and B)). However it is possible that instead, a thin coating of the bridge is left behind on the plate and on the sphere such that the contact is pinned nearly to its original location (compare Figure 19 A) and C)). In the case where the bridge liquid has a low viscosity (\( \eta^* = 0.005 \) here), such a thin coating may provide excellent lubrication. In the context of Figure 12 A), the lateral extent of the bridge would be larger than \( b \), and the pressure in the bridge would be reduced, both of which would reduce the force needed for the separation. We emphasize that a thin coating of the bridge fluid may be left behind regardless of \( \eta^* \). However when \( \eta^* \gg 1 \), e.g. when the outside fluid is air, this thin coating would not affect the fluid
mechanics in the bridge region or separation force. It is only when the bridge has low viscosity that this thin coating affects the separation force.

Figure 19. Liquid bridge at A) initial configuration, and B) after some separation assuming that the contact line recedes along both solid surfaces. C) A thin coating of the bridge liquid remains on the sphere and the plate. Note that in C, the separation distance is identical to that in B, but the contact line on the sphere and the plate is still at its initial value in A.
5.5 Conclusion

Here, we examine the dynamics of a liquid bridge between a sphere and a flat plate being separated from each other. The separation velocities are sufficiently large that viscous forces contribute significantly to the force required for separation. Unlike previous research in this area, this paper focuses on situations where the viscosity of the bridge is far lower than of the external medium within which the particle and the plate are immersed. For the general case of a viscosity mismatch between the bridge fluid and the external fluid, under the lubrication approximation, we develop a theoretical model for the viscous force during separation. The viscous force has contributions from both the pressure outside the liquid bridge (proportional to the viscosity of the outside fluid) and from the pressure inside the liquid bridge. In the case of interest here, when the bridge viscosity is low, the model predicts that the pressure in the bridge is nearly constant and has a value that is governed entirely by the external fluid. Crucially, the bridge region is predicted to contribute to the force even if the bridge fluid is inviscid. Further, as may be expected intuitively, a low viscosity meniscus is expected to reduce the separation force as compared to both a bridge of matched viscosity or separation without a liquid bridge.

The model is compared with experiments in which a sphere is separated from a flat plate with separation distances ranging from a few percent of the sphere radius to equal to the sphere radius. Experiments confirm that a low-viscosity bridge reduces the total force during separation. The magnitude of reduction is more severe at small separations, and at large bridge volumes. The magnitude of the reduction is also almost two-fold larger than predicted by the model. We propose that the reason for this is that a thin layer of the bridge fluid is left behind on the particle and the plate, and this layer serves as a lubricating layer – an effect that is not captured in the model. Finally, experiments also report that the rupture distance increases by as much as two-fold over
the static rupture distance, as the separation velocity increases. Although experiments are conducted and the theory is derived for bridges between a plate and a spherical particle, all results are expected to apply for bridges between a pair of particles as well.
6.0 Modeling of a Low Viscosity Meniscus Immersed in a Higher Viscosity Liquid

Shan-Chen’s implementation of a multicomponent [90] LBM introduces an inter-particle interaction between components. In this method, the inter-particle interaction is repulsive and induces phase separation with a resulting interface between the immiscible components. This method has been used widely to simulate two-component phenomena. For example, the displacement of immiscible fluid in porous media was investigated by Pan et al. [91]. Also, Huang et al. [92] studied the contact angle as a function of different adhesion parameters and proposed an approximation to predict the contact angle. Yang et al. [93] study the interaction between a freely moving particle with different wettability and a freely moving droplet using the Shan-Chen multicomponent LBM method. They reported two types of regimes: agglomeration and separation. In addition, they find that the wettability of the particles is critical in both regimes. Shinto et al. [94] use the LBM method to examine capillary forces in two cases: lateral capillary force between two horizontal cylinders constrained in a liquid-vapor interface and capillary bridge force between two wetting cylinders inside a liquid film covered by vapor. They concluded that LBM reproduces the capillary forces and considers the solid's wettability property.

In this chapter, we first examine three test case scenarios using the Shan-Chen LMB method: surface tension of a droplet in a second fluid, deformation of a droplet, and contact angle as a function of different adhesion parameters. Afterward, this multicomponent LBM-DEM model is used to investigate low viscosity liquid bridge rupture in a more viscous surrounding fluid and, where possible, these results are compared with experiment.
6.1 Multiphase Multicomponent LBM

The Shan-Chen (1993) method is used in this section to model a multicomponent multiphase (MCMP SC) system in three dimensions. In this model, each component has its own distribution function and satisfies the Lattice Boltzmann equation:

$$f_i^\sigma(x + c_i \Delta t, t + \Delta t) = f_i^\sigma(x, t) - \frac{\Delta t}{\tau_\sigma} \left(f_i^\sigma(x, t) - f_i^\sigma, eq(\rho, u)\right). \quad (6.1)$$

where $f_i^\sigma$ is the distribution function of the component $\sigma$. Each component has a relaxation parameter $\tau_\sigma$ that controls the kinematic viscosity of the component $v_\sigma = c^2(\tau_\sigma - 0.5\Delta t)$. In this method the equilibrium distribution function $f_i^\sigma, eq$ can be determined by:

$$f_i^\sigma, eq = w_i \rho \left(1 - \frac{3}{2c^2} u_i^{eq^2} + \frac{3}{c^2} (c_i \cdot u_i^{eq}) + \frac{9}{2c^2} (c_i \cdot u_i^{eq})^2\right). \quad (6.2)$$

The equation 6.2 is similar to the equation 2.6 used for a single component. The only difference is the macroscopic velocity $u_i^{eq}$ that can be calculated by

$$u_i^{eq} = u' + \frac{\tau_\sigma F_\sigma}{\rho_\sigma}, \quad (6.3)$$

where $u'$ is the total velocity of the various components and is defined as

$$u' = \sum_\sigma \left(\sum_i \frac{f_i^\sigma c_i}{\tau_\sigma}\right) / \sum_\sigma \tau_\sigma. \quad (6.4)$$

In equation 6.3 $F_\sigma = F_{ads,\sigma} + F_{c,\sigma}$. $F_\sigma$ is the force acting on the component $\sigma$, that include the fluid-solid adhesion force $F_{ads,\sigma}$ and the fluid-fluid cohesion force $F_{c,\sigma}$. The adhesion force is defined as [93]

$$F_{ads,\sigma}(x,t) = -G_{ads,\sigma}(x,t) \sum_i w_i s(x + c_i \Delta t) c_i, \quad (6.5)$$
where \( s(\mathbf{x} + \mathbf{c}_i \Delta t) \) is a function that is equal to 1 if the neighbor node is a solid node and equal to 0 if the neighbor node is a fluid node. The parameter \( G_{ads,\sigma} \) controls the strength of the adhesion force between fluid-solid. Different groups [91-92] concluded that a negative \( G_{ads,\sigma} \) will result in a wetting fluid, and a positive value will result in a nonwetting fluid.

The cohesion force can be calculated using [93]

\[
\mathbf{F}_{c,\sigma} = -G_{c,\sigma} \rho(\mathbf{x}, t) \sum_i w_i \rho(\mathbf{x} + \mathbf{c}_i \Delta t) \mathbf{c}_i,
\]

where \( G_{c,\sigma} \) is the parameter that controls cohesion force between component \( \sigma \) with a different component \( \bar{\sigma} \).
6.2 Test Cases

6.2.1 Determination of the Surface Tension

As a first test case to validate our model for a multicomponent system, we place a droplet of one fluid (denoted as fluid 1) in the middle of a continuous other fluid (denoted as fluid 2) in the simulated area with no velocity field applied. The Laplace law gives the difference in pressure between the inside of the dispersed fluid bubble/droplet and the outside fluid, where $\Delta P = 2\sigma / R$. Therefore, the interfacial tension or the fluid-fluid cohesion between two different fluids can be measured using the Laplace law.

After initializing the simulation, the model is run until a steady-state is achieved (defined as no additional changes in the measured simulation results, such as the velocity and density of every node). The amount of fluid in the bubble/droplet was changed, and the difference between the pressure inside the bubble (i.e., at the middle of the bubble) and outside of the bubble (the most far away node from the bubble) was determined at steady state. The pressure for the Shan-Chen model is dependent on the number of dimensions and for a D3Q19 velocity model, the pressure is defined by [95]

$$P = c_s^2 \sum_{\sigma} \rho_{\sigma} + 12c^2 G \rho_1 \rho_2$$

(6.7)

where $c_s = \frac{c}{\sqrt{3}}$ is the sound speed and $c = \frac{\Delta x}{\Delta t}$, G dimensions are $1/\rho$ to be consistent with pressure dimensions (M/(LT^2)).

The results obtained from simulating the pressure withing a series of differently-sized bubbles are shown in Figure 20. These results exhibit a linear relationship between the difference
in pressure and the inverse of the radius of the bubble \( (1/R \text{ such that } \Delta P \propto 1/R) \). This figure shows that the results from our model (using the Shan-Chen method) accurately captures the fluid-fluid interaction between two fluid components as the pressure difference follows the Laplace law and the fluid-fluid interfacial tension is captured correctly. For \( G_{\text{coh}} = 1.8 \), the slope is 1.37, which can be used to determine the interfacial tension between the two fluids to \( \sigma = 0.685 \text{ M/T}^2 \) (LB units).

Figure 20. Laplace law test result.
6.2.2 Deformation of a Droplet Under a Shear Flow

In this test, we use our LBM model in combination with the Shan-Chen model to investigate the deformation of a droplet in a shear flow. Periodic boundary conditions were used in each direction (x, y, and z), and the dimension of our domain are $N_x=100$, $N_y=150$, and $N_z=100$. A bubble is located in the middle of the domain with a density of fluid one ($\rho_{\text{fluid1}}$) equal to 2.0 (dimensionless) and a density of fluid two ($\rho_{\text{fluid2}}$) equal to 0.02, so that it is effectively a bubble of pure fluid one. A shear velocity in the x-direction is applied as a function of height with Eq. 6.8.

$$V_{\text{shear}} = \frac{0.5N_y - y}{0.5N_y} V_{\text{shear max}}$$

(6.8)

where $V_{\text{shear max}}$ is the maximum shear velocity, and $y$ is the height in dimensionless lattice units.

The bubble diameter is set to 30 nodes (as seen in Figure 21). Outside the bubble the values of density are inverted to mimic a continuous phase that is nearly pure in fluid two so that $\rho_{\text{fluid1}}=0.02$ and $\rho_{\text{fluid2}}=2.0$, respectively. A value of $G_c = 1.5$ and relaxation parameter $\tau =1$ are set for this test. Once again, the simulations is run until a steady-state is achieved or until the breaking of the droplet is observed.
Figure 21. Initial condition before applying a shear velocity.

Figure 22 shows a concentration plot where each node is color-coded with respect to the amount of fluid one present at that particularly location such that a node is orange if $\rho_{\text{fluid1}} \geq 0.1$ and yellow when $\rho_{\text{fluid1}} < 0.1$. The results are presented as a 2-D plane located at the center of the 3-D simulation. The bubble remains intact if the shear velocity is small (i.e., there is no deformation, see Figure 22 A)). Under moderate shear velocity, the bubble at steady-state has a deformation with an elliptical shape (Figure 22 B)). At high shear velocity, the bubble breaks, and the two-fluid components are mixed; this was the case in Figure 22 C), in which the simulation never achieved a steady state. The deformation of the bubble depends on the interfacial tension and the shear velocity. An increase in shear velocity will increase the deformation of the bubble,
and when it is high enough, the bubble breaks up. Interfacial tension can be tuned between fluids by changing the value of $G_c$.

Figure 22. Droplet density at different shear velocities: a) 0.000001, b) 0.000015, and c) 0.0001 (dimensionless).
6.2.3 Determining the Contact Angle

Our final validation simulation series is aimed at testing whether the fluid-solid adhesion interaction is captured correctly with the model and whether the interaction can be tuned through judicious choice of interaction parameter(s). The test simulations are initialized with a rectangle of fluid one which is placed near a solid wall and surrounded by a continuous phase of fluid two. As before, the density in the rectangle for fluid one was set to 2.0 and 0.06 for fluid two. Similarly, outside the rectangle the density of fluid one is set to 0.06 and 2.0 for fluid 2. The contact angle is determined as a function of $G_{\text{ads}}$, and in this series of tests we take $G_{\text{ads}1} = -G_{\text{ads}2}$. Similar to the previous simulations, when a steady-state is achieved, the simulations are stopped. The results of these trials are shown in Figure 23 (once again as a 2-D representation of the middle plane of the 3D simulation). The contact angle ($\theta$) is related to the surface tension and interfacial tension of the two fluids and can be calculated using Young’s equation (Eq. 6.9)

$$\cos \theta = \frac{\sigma_2 - \sigma_1}{\gamma}$$  \hspace{1cm} (6.9)

where $\sigma_2$ and $\sigma_1$ are the surface tension of fluid 2 and 1, respectively and $\gamma$ is the interfacial tension. Young’s equation applied to the Shan-Chen multicomponent model is approximated by the following equation (Eq. 6.10), where the contact angle is a function of $G_{\text{ads}}$ and $G_c$ [92]

$$\cos \theta = \frac{G_{\text{ads}2} - G_{\text{ads}1}}{G_c \frac{\rho_1 - \rho_2}{2}}$$  \hspace{1cm} (6.10)

We observe that, when the value of $G_{\text{ads}}$ is negative, the fluid wets the surface ($\theta < 90^0$), and when it is positive, the fluid is nonwetting ($\theta > 90^0$), matching what has been reported previously in the literature [92, 96]. In Table 4, we show a listing of the obtained contact angle using Eq. 6.10 as a function of $G_{\text{ads}}$, and compare this to the contact angle that has been measured
from the simulated images. As can be seen, the contact angles measured from the model are in
good agreement with the contact angles predicted from Eq. 6.10; therefore, we conclude that our
model can capture the wettability properties of a pair of fluids (with a surface), and that this
wettability can be controlled with the value of $G_{\text{ads}}$. 
### Table 4. Contact angle as a function of $G_{ads}$

<table>
<thead>
<tr>
<th>Case</th>
<th>$G_{ads}$</th>
<th>Approximation of contact angle $\theta$</th>
<th>Measured contact angle $\theta$</th>
</tr>
</thead>
<tbody>
<tr>
<td>A</td>
<td>-0.1</td>
<td>80.3°</td>
<td>82.3°</td>
</tr>
<tr>
<td>B</td>
<td>-0.3</td>
<td>59.7°</td>
<td>66.3°</td>
</tr>
<tr>
<td>C</td>
<td>-0.5</td>
<td>32.7°</td>
<td>35.4°</td>
</tr>
<tr>
<td>D</td>
<td>0.1</td>
<td>99.7°</td>
<td>99.2°</td>
</tr>
<tr>
<td>E</td>
<td>0.3</td>
<td>120.3°</td>
<td>117.2°</td>
</tr>
<tr>
<td>F</td>
<td>0.5</td>
<td>147.3°</td>
<td>150.3°</td>
</tr>
</tbody>
</table>

Figure 23. Contact angle at different $G_{ads}$: A) -0.1, B) -0.3, C) -0.5, D) 0.1, E) 0.3, and F) 0.5
6.3 Low Viscosity Liquid Bridge in a Viscous Surrounding Fluid Simulation

Here we report how the LBM-DEM multicomponent model was used to investigate the rupture of a liquid bridge. In both experiments and simulations, we create a liquid bridge between a particle and a flat wall and investigate the resultant forces on the particle. Three cases were studied: a particle near a flat wall with no secondary fluid (i.e., no liquid bridge, which we will refer to as “no meniscus”), a case where the liquid bridge (meniscus) is comprised of a fluid with a lower viscosity than that of the surrounding fluid, and a case where the viscosity of the surrounding fluid is matched to that of the meniscus fluid. In cases with differing viscosities, a viscosity ratio of 10 was investigated as a greater viscosity ratio produces instability in our simulations. In contrast to the comparison experimental procedure, in the simulations the particles move at a constant separation velocity away from the flat wall rather than moving the flat wall away from the particle, as is done in the experiments. Because of this difference, at large separation distances, we observe simulated forces that represent the force from particle drag (as opposed to no net force as observed in the companion experiments).

6.3.1 Theory

The force involved in this simulation is similar to the force involved in Chapter 5. Specifically, we anticipate a combination of viscous and capillary forces to act on the particle. As mentioned, the main difference between the companion experiments and the simulations is that in the simulations the particle moves at a constant velocity instead of moving the wall away from a stationary particle (as is done in the experiments). As such, we need to consider the drag force which is given by
\[ F_{\text{drag}} = \frac{1}{2} C_d \rho A v^2 \]  

(6.11)

where \( A \) is the cross-sectional area of a sphere \((A = \pi r^2)\) and \( C_d \) is the drag coefficient Eq. 6.12 [97].

\[ C_d = \frac{24}{Re} \]  

(6.12)

In this equation \( Re \) is the Reynold’s number and represents the ratio of inertial forces to viscosity forces and is given by

\[ Re = \frac{\rho v d}{\eta} \]  

(6.13)

In the cases where there is no meniscus, the resulting forces on the moving particle include the viscous force from Eq. (5.3) and the drag force outlined above so that we obtain

\[ F_{\text{tot}} = F_{\text{visc}} + F_{\text{drag}} = 6\pi \eta R^2 \frac{1}{S_0} v + \frac{1}{2} C_d \rho A v^2 \]  

(6.14)

The other cases investigated here include the force from the meniscus. Thus, they include the capillary force (Eq. 5.14) in addition to the viscous forces from the meniscus, the viscous forces from the surrounding fluid (Eq. 5.12) and the drag force. This combination of forces is given by

\[ F_{\text{tot}} = \frac{4\pi R \gamma \cos \theta}{1 + S_0 \sqrt{\frac{\pi R}{V}}} + \frac{6\pi \eta_0 R^2 v}{S_0} \left( -\frac{1}{\sqrt{V^*} + 1} + \frac{\left(\sqrt{V^*} + 1\right) - 1}{V^* + 1} \right) \]  

(6.15)

\[ + \frac{1}{2} C_d \rho A v^2 \]
To have a more direct comparison between the experiments and the simulations the simulation force reported in this chapter $F_{\text{sim}}$ is the force of the LBM-DEM model $F_{\text{model}}$ minus the drag force $F_{\text{drag}}$.

$$F_{\text{sim}} = F_{\text{model}} - \frac{1}{2} C_d \rho A v^2$$

(6.16)

### 6.3.2 Simulation’s Parameters

The simulations that include a fluid meniscus are initialized with a rectangle of fluid one placed between the particle and the wall. We use an initial separation distance between the sphere and the wall of 102 microns which corresponds to one lattice node spacing (Figure 24). The density in the rectangle for fluid one is set to 2.0 and to 0.02 for fluid two. Outside the rectangle, the density of fluid one is set to 0.02 and to 2.0 for fluid 2. The particle diameter in these simulations is 1.0 mm, and the meniscus volume is approximately 2.5 $\mu$L. The relationship diameter to volume is equal to the 5 $\mu$L meniscus case investigated experimentally in Chapter 5. The adhesion parameter $G_{\text{ads}}$ is set to $G_{\text{ads1}} = -0.5$ and $G_{\text{ads2}} = 0.5$ which yields an effective contact angle equal to 32.67$^\circ$. The viscosity and the interfacial tension (measured using the Laplace law) of each simulated fluid are shown in Table 5. For the cases that include no meniscus only one fluid is simulated (i.e., there is no interstitial rectangle of fluid), but all other operational parameters are kept constant. Similar to what is done in the experimental work outlined in Chapter 5, we investigate the effect of velocity and meniscus volume on the total force exerted on a particle during the rupture process. We perform simulations both with and without a meniscus at different separation velocities: $v = 1280, 2560, \text{and } 3840 \mu \text{m/s}$. The velocity range was selected to be higher
than the experimental separation velocity because the viscosity ratio in the simulations is 10 as compared to a ratio of 78 as is used in the experimental results.

### Table 5. Simulated fluid properties.

<table>
<thead>
<tr>
<th>Liquids</th>
<th>Viscosity (Pa s)</th>
<th>Interfacial tension (mN/m)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Surrounding fluid</td>
<td>0.0039</td>
<td>---</td>
</tr>
<tr>
<td>Low viscosity meniscus</td>
<td>0.00039</td>
<td>9.3x10^-4</td>
</tr>
<tr>
<td>Viscosity matched meniscus</td>
<td>0.0039</td>
<td>2.4x10^-4</td>
</tr>
</tbody>
</table>

Figure 24. Simulation initial conditions with a liquid bridge.
6.3.3 Results

6.3.3.1 Force Without Meniscus

The total force exerted on the particle for the no meniscus case is presented in Figure 25 and is compared to the theoretical force for this case as calculated using Eq. 6.14 (without drag force). The viscous is a function of velocity and the total force increases with an increase in velocity. The viscous force depends on separation and at long distances is close to $F_{\text{Visc}} \sim 0$. As such, when using Eq. 6.14 for the total force in the case with no meniscus we observe a good agreement with the simulated results. Figure 25 B) shows the ratio of the total force to the velocity in the case with no meniscus and, as one would expect from the model, we observe a linear relationship with velocity ($F_{\text{tot}} \propto v$). From these results, we conclude that our model can capture the velocity dependence the viscous forces as well as how these forces vary with the distance from the surface.
Figure 25. A) Total force for no meniscus compared to the theoretical force Eq. 14 (without drag force). B) Ratio of the force and velocity.
6.3.3.2 Meniscus Evolution

The evolution of the meniscus at two different particle velocities: \( \nu = 1280 \) and \( 3840 \) \( \mu \text{m/s} \) are presented in Figure 26 A-D) and E-F), respectively. At high separation velocity, the meniscus is fatter than at a lower separation velocity and this thickness tendency is very noticeable when Figures 26 C) and G) are compared. The rupture distance increases with an increase in separation velocity as shown in Figure 26 C) and H), where at \( 1280 \) \( \mu \text{m/s} \), the liquid bridge is broken at \( S_0/R = 3.0 \), while at \( 3840 \) \( \mu \text{m/s} \) the bridge is broken near to \( S_0/R = 3.5 \). Both of these simulation trends were also observed in the experiments that are shown in Figure 14: a fatter meniscus at high velocity and an increase in the rupture distance with an increase in separation velocity. This comparison is qualitative since the viscosities and the viscosity ratio are different in the simulations and the experiments.

![Figure 26. Meniscus evolution at different separation distances (labeled in the top row). A)-D) with a separation velocity \( \nu = 1280 \mu \text{m/s} \) and E)-H) 3840 \( \mu \text{m/s} \).](image)
6.3.3.3 Effect of Meniscus Viscosity on the Force

As shown in Eqn. 6.15, the total force during separation experienced by a particle with a liquid bridge in a surrounding fluid is a function of the separation velocity, liquid bridge and surrounding fluid viscosity, and interfacial tension. Figure 27 compares the total force with a separation velocity of $v = 2560 \mu m/s$ for three cases: no meniscus, a viscosity-matched meniscus, and a low viscosity meniscus. Also, the theoretical force for the no meniscus case is included.

A low viscosity meniscus decreases the total force when compared to a case that has no meniscus. In contrast, the viscosity-matched meniscus shows a slightly higher total force than the no meniscus. The viscous force is the same in both the no meniscus and the viscosity-matched meniscus cases since both cases involve fluids that have the same viscosity; however, in the viscosity-matched meniscus case, the addition of the capillary force increases the total force, resulting in a higher overall value. The same trend in the relative magnitude of the forces in the three comparator cases – i.e., with a low viscosity meniscus, a viscosity-matched meniscus, and no meniscus -- was observed experimentally in Chapter 5. It should be noted that, in the simulation results, a fluctuation is observed in the measurement of the net force. This fluctuation in the force is caused because the particle's velocity is high and the simulation is confined to a bounded system, thus the motion of the particle produces a velocity field in the simulated area (which causes a small additional (fluctuating) force).
Figure 27. Total force for a low viscosity and a viscosity match meniscus with a volume of 2.5 \( \mu \text{L} \), and no meniscus at a separation velocity of \( v = 2560 \ \mu \text{m/s} \), and the theoretical value of the no meniscus using Eq. 6.14 and 6.15 (without drag force).
6.3.3.4 Velocity Effect

Figure 28 compares the total simulated force for the lower-viscosity meniscus and no meniscus cases at A) $v = 1280 \, \mu\text{m/s}$ and B) $3840 \, \mu\text{m/s}$. The total forces for the no meniscus cases were consistently higher than the lower-viscosity meniscus case at the separation velocity range investigated. Comparing the cases of the (low viscosity) meniscus and no meniscus at short distances of separation, the effective force is higher in the case with no meniscus. At a long separation distance in the case of a $v = 1280 \, \mu\text{m/s}$ separation velocity (i.e., for $S_0/R > 2.0$), one notes that the total force for the low-viscosity meniscus case becomes comparable to, or slightly larger than the case with no meniscus. In the case of a $v = 3840 \, \mu\text{m/s}$ separation velocity, the viscous force is more relevant over a longer distance thus the forces for the competing cases do not become comparable until a longer distance (i.e., $S_0/R > 3$). Once again, these results are comparable to the experimental result obtained with a viscosity ratio of 78 in Chapter 5.
Figure 28. Total simulated force with meniscus and no meniscus at A) $v = 1280 \, \mu m/s$ and B) $3840 \, \mu m/s$ (without drag force).
The ratio of the total forces to the velocity for the low-viscosity meniscus case is shown in Figure 29. Interestingly, we can see that we do not observe a collapse of the data (indicative of a linear relationship between the total force and the separation velocity) for all velocities studied. This suggests that our results are in the capillary regime at a separation velocity of $v = 1280 \, \mu m/s$, but that the results from the separation velocities of $v = 2560 \, \mu m/s$ and higher are likely within the viscous-dominated regime. These results follow the same pattern observed experimentally in Chapter 5 in which results that were obtained for experiments above a minimum separation velocity, the total force was proportional to the velocity.

![Figure 29. Ratio of the total forces and velocity for the low viscosity meniscus.](image-url)
Figure 30 shows the total force for the low viscosity meniscus is compared to the theoretical force obtained from Eq. 6.15 as a function of different separation velocities. For all the velocities investigated, the total force calculated from the theory overestimates the result obtained in the simulated results. More specifically, the viscous force contribution is overestimated by roughly a factor of 2. Interestingly, this tendency to overestimate matches a similar trend of overestimation observed when comparing the experimental results in Chapter 5 to the same theory. As such, the same rationale that was discussed in more detail in that chapter is relevant here.

![Graph showing total force for the meniscus at different velocities compared to Eq. 6.15 (without drag force).](image)

**Figure 30. Total force for the meniscus at different velocities compared to Eq. 6.15 (without drag force).**

### 6.3.3.5 Effect of Meniscus Volume

The effect of meniscus volume on the total force was investigated using two different volumes, $V = 2.5$ and $4 \mu$L, respectively. The results displayed in Figure 31 show that the initial force is lower for the case with a higher bridge volume at very small separations. This can be understood because the area covered by the low-viscosity meniscus is larger with increasing bridge...
volume, thus the viscous force contribution of the surrounding fluid decreases with increasing (low viscosity) bridge volume. Similarly, it can be understood that this difference in force between the two meniscus volumes is only present at a short separation distance since the rapid thinning of the bridge obviates the increase in area rather quickly. As such, once the separation reached $S_0/R > 0.7$, the observed force is equal for both meniscus/bridge volumes. As with all other simulated results, these qualitative patterns were observed experimentally in our results reported in Chapter 5.

Figure 31. The total force exerted in a particle with a low viscosity bridge with a volume of $V = 2.5 \, \mu\text{L}, 4 \, \mu\text{L}$ and no meniscus with $v = 2560 \, \mu\text{m/s}$ (without drag force).
In this chapter, we develop a multicomponent model and validate it with three different tests. We demonstrated that the model could be tuned to capture solid wettability properties. Also, we show that this model correctly captures the fluid-fluid interaction between two fluid components. We then used this model to investigate differences in the observed force for cases that include no bridge as well as the cases of rupture of a viscosity-matched bridge and a low viscosity bridge (embedded within a more viscous continuous fluid). The results from the simulations were qualitatively compared with experimental results. The simulation results matched the patterns observed experimentally (as reported in Chapter 5). Finally, it is noted that the total force is reduced with a low viscosity meniscus, and at high velocity, the meniscus force was proportional to the velocity.
7.0 Summary and Outlooks

Particle-particle and particle-fluid interactions are essential in many of the industries. These interactions can be complicated and are widely affected by the fluid's viscosity. A combination of experiments and simulation can be helpful to understand these interactions in different processes. This dissertation studied two problems: the mixing in the Ram mixer and the liquid bridge rupture in a viscous continuum fluid with experiments and simulations. In Chapter 3, we developed an LBM-DEM model capable of capturing particle-particle interactions and fluid-particle interactions and the effect of viscosity in these interactions. Chapter 4 explores the RAM mixer's mixing performance with experiments and simulations. Chapter 5 investigated the low viscosity meniscus rupture inside a viscous continuum fluid. Finally, Chapter 6 develops a multicomponent model that accurately captures adhesion and cohesion force. We used this method to investigate low viscosity meniscus rupture inside a viscous fluid.

The modeling work in this dissertation was carried out using LBM coupled with DEM considering the momentum transport between solids and liquids. The LBM method is capable of recovering the Navier-Stokes equation. The DEM determines the particle's positions, velocity, and orientation, considering all the forces acting in each particle. Our LBM-DEM is capable of capturing interfacial tension between two fluids and wettability properties.
7.1 Mixing in RAM

The mixing of solids and liquids is relevant in different industries. Different parameters can affect the mixing performance: mixer types, fluid properties, and particles properties (density, shape, size). In chapter 4, we evaluated the mixing performance of the RAM mixer in a slurry with high viscosity. We determine the main limitations of this type of mixer experimentally and with simulations. The size of the particles was important, and when the small particles have a low concentration, we notice segregation toward the bottom with a bad mixing performance. The acceleration was an important parameter, and at high viscosity, the acceleration improved the mixing performance. The mixing mechanism at high viscosity was determined, in which the mixing occurs when the temperature increase and the dynamic viscosity decreases. Without this temperature change, the mixing performance was poor. The mixing performance was determined experimentally and with simulations, and the same patterns were observed.

Future outlook in exploring the RAM mixing performance can investigate the size effect for smaller particles. To determine if segregation toward the bottom is not dependent on the size of particles. This will help determine if the RAM can mix small particles in low concentrations in different sizes. Another exciting factor that can be investigated is density segregation, where usually the heavy particles are concentrated in the bottom.
7.2 Low Viscosity Liquid Bridge

Capillary force in a liquid bridge between two solids is relevant across multiple chemical processes. Most of the model and experimental studies of liquid bridges available in the literature are about viscous liquid bridges, usually in the air or in a less viscous continuum fluid. In Chapters 5 and 6, we investigated the rupture of a low viscosity meniscus inside a viscous fluid. The force obtained results clearly show that a low viscosity meniscus decreases the total force compared to the no meniscus case and a viscosity match meniscus. Therefore, less force is needed to break the liquid bridge when compared to a high viscosity liquid bridge. The force is directly proportional to the velocity for the low viscosity meniscus at high velocity because the viscous force dominates the capillary force. Similar to the velocity-dependent observed in the no meniscus cases or an infinite liquid film. The force reduction was a function of the low viscosity meniscus volume, where an increase in meniscus volume decreases the total force. Rupture distance increases when the separation velocity increases, similar to a viscous liquid bridge in air.

An area that can be explored with our LBM-DEM multicomponent model is under which conditions a freely moving liquid bridge between two particles will break and under which conditions the bridge remains. This type of experiment is challenging since the initial condition of a liquid bridge in a continuum phase is hard to replicate, and it is hard to do measurements under shear flow. Therefore, our LBM-DEM multicomponent can be used to investigate the liquid bridge under shear and elongated flow. These results will be helpful to develop a curve similar to the "Grace curve" applied to a liquid bridge between particles.


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