Development and Demonstration of a General-Purpose Model for Brownian Motion

THESIS

Presented in Partial Fulfillment of the Requirements for the Degree Master of Science in the Graduate School of The Ohio State University

By

Derek Endres B.S. (Honors)

Graduate Program in Mechanical Engineering

The Ohio State University

2011

Master's Examination Committee:

Dr. Sandip Mazumder, Advisor

Dr. Shaurya Prakash
Copyright by

Derek Joseph Endres

2011
Abstract

Brownian motion is an important phenomena demonstrated by sub-micron sized particles in fluids. The objective of the current work is to develop a general-purpose numerical model to simulate Brownian motion and explore it for various applications of practical interest. To accomplish this goal, a model based on the Langevin equation was developed and implemented. The resulting stochastic ordinary differential equations were solved using the Newton-Raphson method after backward Euler discretization. In order to track the particles through the background fluid, a three-dimensional (3-D) tracking algorithm was developed. The solver is applicable to arbitrary two-dimensional (2-D) and 3-D geometries discretized using unstructured mesh topology. The Brownian motion solver was fully coupled to the carrier fluid flow solver, enabling unsteady flows to be simulated. In order to apply the model to various applications, viscous drag, Brownian forces, lift, and thermophoretic forces were considered. These forces all contribute to the particle’s velocity and resulting motion. The Brownian motion model was first validated against analytical results and then explored for various applications where Brownian motion is important. From the simulations, it is shown that Brownian motion is increased by decreasing the particle size, increasing the fluid temperature, or decreasing the fluid viscosity. One application where Brownian motion is taken advantage of is in micro-scale filters. By simulating micro-scale filters, they can be optimized so that capture efficiencies can
be improved. It is shown by performing simulations under different operating conditions, that the capture efficiency of micro-filters can be increased for a given particle diameter by increasing fluid temperature or decreasing fluid viscosity. Another application investigated in this work is the chemical vapor deposition (CVD) of Aluminum Nitride (AlN). During this process, it has been experimentally observed that particles are formed as suspended solids in the gas above the epitaxial film. Numerical simulations are conducted to shed light on the trajectory of these sub-micron particles as they travel through the reactor. Thermophoretic forces are found to be the dominant forces acting on such particles in steady CVD, and as experimentally observed; these forces prevent the AlN particles from getting incorporated in the epitaxial film. It is shown that Brownian motion has a much more significant contribution to the trajectory of formed AlN particles in pulsed AlN CVD than steady AlN CVD. In summary the main contributions of the thesis are as follows:

• Development of a fully coupled 3-D Brownian motion model based on the Langevin equation along with a particle tracking algorithm that is applicable to any mesh topology, including unstructured meshes.
• Simulation-based optimization of H-filters and cylindrical filters.
• Simulation-based understanding of the fate of AlN particle within CVD reactors of various configurations and under various operating conditions.
This document is dedicated to my family.
Acknowledgements

I would like to thank my advisor, Professor Sandip Mazumder, for helping me along every step of the way in this research. I thank him for his patience and guidance during the course of my research. Without his help and guidance none of this would have been accomplished. I am grateful to Professor Shaurya Prakash for serving on my thesis defense committee and his valuable help during my defense. Thanks to the entire lab group from over the past 2 years for all their help and support. I also would like to thank my fiancé Patti Wernke for helping me with this thesis and for all encouragement and support of my goal of getting a Master’s of Science in mechanical engineering. This research was supported in part by a Graduate Laboratory Teaching position by The Ohio State College of Engineering through the Mechanical Engineering department. ESI Group, North America, is acknowledged for providing license of CFD-ACE+tm.

A final, special thanks goes out to all of my friends and family, especially those who helped me with this thesis. Without their loving support, I would not be the person I am today.
Vita

June 2005 ............................................................. St. Xavier High School

March 2010 ........................................................ B.S. Mechanical Engineering, The Ohio State University

March 2010 to present .......................................... Graduate Teaching Associate, Department of Mechanical Engineering, The Ohio State University

Publications


“Numerical Investigation of Pulsed Chemical Vapor Deposition of Aluminum Nitride to Reduce Particle Formation”, Derek Endres and Sandip Mazumder, *Journal of Crystal Growth* (accepted)

Fields of Study

Major Field: Mechanical Engineering
# Table of Contents

ABSTRACT ................................................................................................................................. II

ACKNOWLEDGEMENTS .............................................................................................................. V

VITA ............................................................................................................................................... VI

TABLE OF CONTENTS .................................................................................................................. VII

LIST OF FIGURES ......................................................................................................................... IX

LIST OF TABLES .......................................................................................................................... XIII

NOMENCLATURE ......................................................................................................................... XIV

1 INTRODUCTION ........................................................................................................................ 1

1.1 HISTORY AND DESCRIPTION OF BROWNIAN MOTION ......................................................... 1

1.2 APPLICATIONS OF BROWNIAN MOTION ........................................................................... 5

1.3 OBJECTIVES ......................................................................................................................... 10

1.4 ORGANIZATION OF THESIS ............................................................................................... 11

2 MATHEMATICAL MODEL AND SOLUTION ............................................................................ 13

2.1 EQUATIONS OF MOTION OF BROWNIAN PARTICLE ......................................................... 13

2.2 GOVERNING EQUATIONS FOR CARRIER FLUID ................................................................ 17

2.3 SOLUTION PROCEDURE ....................................................................................................... 18

2.3.1 Imported Data for Brownian Motion Solver ................................................................... 20
# List of Figures

| Figure 1: Trajectories of 3 Different Particles Exhibiting Brownian Motion as Observed by Perrin (Perrin, 1916) ....................................................................................................................................................... 3 |
|--------------------------------------------------|---------------------------------------------------------------------------------------------------------|
| Figure 2: Micro-Cylindrical Filter (Jiang, et al., 2005) .................................................................................................................. 7 |
| Figure 3: H-filter Schematic (Holl et. al., 1996) .......................................................................................................................... 8 |
| Figure 4: Schematic of MOVPE of AlN (Endres, 2010) ..................................................................................................................... 10 |
| Figure 5: Flow Chart Depicting Overall Model ............................................................................................................................ 19 |
| Figure 6: Flow Chart Depicting Carrier Fluid Solution .............................................................................................................. 19 |
| Figure 7: Flow Chart Depicting Brownian Particle Solver ........................................................................................................... 20 |
| Figure 8: Flow Chart Depicting Brownian Particle Equation Solver ............................................................................................................ 24 |
| Figure 9: Flow Chart Depicting ODE Solver .................................................................................................................................. 28 |
| Figure 10: Particle Tracing Schematic ........................................................................................................................................ 31 |
| Figure 11: Flow Chart Depicting Particle Tracking Algorithm .................................................................................................. 32 |
| Figure 12: Intersect 2-D Algorithm Schematic ............................................................................................................................. 35 |
| Figure 13: CFD-GEOM Sample Screen Shot .................................................................................................................................... 36 |
| Figure 14: CFD-ACE-GUI Sample Screen Shot .............................................................................................................................. 37 |
| Figure 15: CFD-ACE Sample Residual Plot .................................................................................................................................... 38 |
| Figure 16: CFD-VIEW Sample Screen Shot .................................................................................................................................... 39 |
| Figure 17: Sample 0.05 µm Diameter Particle Trajectory for a Time Period of 0.01 s in a 2-D Steady Flow with a Reynolds Number of 400 .......................................................................................................................... 42 |

---

ix
Figure 18: Sample 0.05 µm Diameter Particle’s X and Y-Velocities for a Time Duration of 0.01 s in a 2-D Steady Flow with a Reynolds Number of 400

Figure 19: Sample 0.05 µm Diameter Particle’s Brownian Forces Experienced for a Time Duration of 0.01 s in a 2-D Steady Flow with a Reynolds Number of 400

Figure 20: 0.05 µm Diameter Particle Trajectory in the X-Direction for a Time Duration of 0.01 s in a Flow with a Reynolds Number of 400 for a 2-D Steady Simulation with 1000 Particles

Figure 21: 0.05 µm Diameter Particle Trajectory in the Y-Direction for a Time Duration of 0.01 s in a Flow with a Reynolds Number of 400 for a 2-D Steady Simulation with 1000 Particles

Figure 22: 0.05 µm Diameter Particle RMS Distance in the Y-Direction for a Time Duration of 8 s in a Stagnant Flow for a 2-D Steady Simulation Based on 5000 Particles

Figure 23: Y-Direction RMS with Varying Particle Diameter for a Time Duration of 20 ms in a Stagnant Flow for a 2-D Simulation Based on 1000 Particles

Figure 24: Y-Direction RMS with Varying Fluid Temperature for a Time Duration of 20 ms in a Stagnant Flow for a 2-D Steady Simulation Based on 1000 Particles

Figure 25: Y-Direction RMS with Varying Fluid Viscosity for a Time Duration of 20 ms in a Stagnant Flow for a 2-D Steady Simulation Based on 1000 Particles

Figure 26: X-Direction RMS with a Time Step of 0.2 s and 0.04 s for a Time Duration of 1 s for a 3-D Simulation Based on 1000 Particles

Figure 27: 2-D Schematic for 3-D Unsteady Fluid Validation

Figure 28: 3-D Unsteady Flow Particle Tracking Validation

Figure 29: Schematic of a Typical H-Filter

Figure 30: H-Filter Velocity Vectors at Top Intersection Point

Figure 31: Large Particle (10 µm) Trajectory in an H-Filter

Figure 32: Small Particle (.1 µm) Trajectory in an H-filter when Filtered

Figure 33: Small Particle (.1 µm) Trajectory in an H-filter when not Filtered

Figure 34: H-Filter Capture Efficiency with Varied Particle Diameters for 500 Particles
Figure 35: H-Filter Capture Efficiency with Varied Fluid Temperature for 500 Particles ..................................... 68
Figure 36: H-Filter Capture Efficiency with Varied Fluid Viscosity for 500 Particles ............................................. 69
Figure 37: Y-Z Plane of Cylindrical Filter Schematic ................................................................................................. 70
Figure 38: 0.1 μm Diameter Particle Trajectory in a 3-D Cylindrical Filter ................................................................. 72
Figure 39: 0.01 μm Diameter Particle Trajectory in a 3-D Cylindrical Filter ................................................................. 73
Figure 40: 3-D Cylindrical Filter Capture Efficiency with Varying Particle Diameters (Wu et. al., 2010) ............... 74
Figure 41: 3-D Cylindrical Filter Capture Efficiency in First Ten Percent of Channel (Wu et. al., 2010) .............. 75
Figure 42: 3-D Cylindrical Filter Capture in Each Section (Wu et. al., 2010) .............................................................. 76
Figure 43: Error Analysis of the 3-D Cylindrical Filter Simulations ............................................................................. 78
Figure 44: 2-D Schematic of a 3-D Horizontal Chemical Reactor ................................................................................. 81
Figure 45: 0.05 μm Diameter Particle Trajectory in a Horizontal Reactor with Thermophoretic Forces Active ................................................................................................................................................. 83
Figure 46: 1 μm Diameter Particle Average Trajectory in a Horizontal Reactor with Thermophoretic Forces Active for 500 Particles ................................................................................................................. 84
Figure 47: Temperature Contour of a Horizontal Reactor ............................................................................................ 85
Figure 48: 1 μm Diameter Particle Trajectory in a Horizontal Reactor with Thermophoretic Forces ..................... 87
Figure 49: Schematic of the AlN Decomposition and Reaction Pathways (Mihopoulos et. al., 1998) ................. 88
Figure 50: AlN Concentration Contour Plot for a Horizontal Reactor ........................................................................ 90
Figure 51: Varied Diameter AlN Particle Trajectories in a Horizontal Reactor .......................................................... 91
Figure 52: Thomas Swan® 2-D Axis-Symmetric Reactor Schematic ........................................................................... 92
Figure 53: Thomas Swan® Temperature Gradient ........................................................................................................ 93
Figure 54: 0.05 μm Diameter AlN Particle Average Trajectory in the Thomas Swan® Reactor ................................. 94
Figure 55: Varied Diameter AlN Particle Average Trajectories in the Thomas Swan® Reactor for 400 Particles .......... 96
Figure 56: 0.1 μm Diameter AlN Particle Average Trajectory in the Thomas Swan® Reactor for 400 Particles ................................................................................................................................................. 97
Figure 57: Mass Flow Rate Schematic for Pulsed CVD .................................................................98

Figure 58: 1 μm Diameter AlN Particle Trajectory in Unsteady Flow: Example 1 ..................100

Figure 59: 1 μm Diameter AlN Particle Trajectory in Unsteady Flow: Example 2 ...............101

Figure 60: 0.1 μm Diameter AlN Particle Formed above Wafer Front Edge Trajectories in Unsteady Flow:
   Example 1 .......................................................................................................................................103

Figure 61: 0.1 μm Diameter AlN Particle Formed above Wafer Front Edge Trajectories in Unsteady Flow:
   Example 2 .......................................................................................................................................104
List of Tables

Table 1: Error Analysis of Overall Filter Capture Efficiency with 10 Simulations of 1000 0.1 μm Particles ..79

Table 2: Kinetic Mechanism for the Growth of AlN (Mihopoulos et. al., 1998) ..................................................89
Nomenclature

A  cell face location
A_f cell face area [m^2]
b linear drag coefficient [N-s/m]
B cell face location
b(t) Brownian force per unit mass [m/s^2]
C_c Cunningham slip correction factor
C_m thermal coefficient
C_s temperature slip constant
C_t momentum accommodation coefficient
C_{\text{Saff}} Saffman lift coefficient
d, d_p particle diameter [m]
d distance particle travels in one time step [m]
D Stokes-Einstein diffusion coefficient [m^2/s]
F_B Brownian force [N]
F_{\text{dr}} drag force [N]
F_{\text{L}} Saffman lift force [N]
F_{\text{t}} thermophoretic force [N]
G_i Gaussian random number with zero mean and unit variance
h enthalpy [J]
J Jacobian
k iteration number
k_b Boltzmann constant [J/K]
k_c thermal conductivity of fluid [W/m-K]
k_d thermal conductivity of particle [W/m-K]
K_n Knudsen number
l distance between cell face intersection point and particle [m]
L characteristic length for Reynolds number calculation [m]
m_p mass of particle [kg]
\mathbf{n}_f cell face normal
O origin
p pressure [Pa]
q arbitrary vector
N_A Avogadro's number [1/mol]
n_c number of fluid particles per unit volume [molecules/m^3]
R universal gas constant [J/mol-K]
<table>
<thead>
<tr>
<th>Symbol</th>
<th>Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>$Re$</td>
<td>Reynolds number</td>
</tr>
<tr>
<td>$r_p$</td>
<td>particle location [m]</td>
</tr>
<tr>
<td>$S$</td>
<td>surface of cell</td>
</tr>
<tr>
<td>$t$</td>
<td>time [s]</td>
</tr>
<tr>
<td>$\Delta t$</td>
<td>time step [s]</td>
</tr>
<tr>
<td>$T$</td>
<td>absolute temperature [T]</td>
</tr>
<tr>
<td>$\nabla T$</td>
<td>temperature gradient [T/m]</td>
</tr>
<tr>
<td>$V$</td>
<td>volume of cell</td>
</tr>
<tr>
<td>$u_c$</td>
<td>velocity of carrier fluid [m/s]</td>
</tr>
<tr>
<td>$u_p$</td>
<td>velocity of particle [m/s]</td>
</tr>
<tr>
<td>$v$</td>
<td>velocity [m/s]</td>
</tr>
<tr>
<td>$x_{rms}$</td>
<td>root mean square displacement in any direction [m]</td>
</tr>
<tr>
<td>$\lambda$</td>
<td>mean free path [m]</td>
</tr>
<tr>
<td>$\sigma$</td>
<td>molecular diameter of carrier fluid molecules [m]</td>
</tr>
<tr>
<td>$\rho_c$</td>
<td>density of carrier fluid [kg/m$^3$]</td>
</tr>
<tr>
<td>$\rho_p$</td>
<td>density of particle [kg/m$^3$]</td>
</tr>
<tr>
<td>$\Phi$</td>
<td>scalar in divergence theorem</td>
</tr>
<tr>
<td>$\nabla \Phi$</td>
<td>gradient of scalar in divergence theorem</td>
</tr>
<tr>
<td>$\Phi_f$</td>
<td>dummy variable face value</td>
</tr>
<tr>
<td>$\mu$</td>
<td>dynamic viscosity of carrier fluid [N-s/m$^2$]</td>
</tr>
<tr>
<td>$\tau$</td>
<td>shear stress [Pa]</td>
</tr>
<tr>
<td>$\omega_c$</td>
<td>local vorticity of the fluid [1/s]</td>
</tr>
<tr>
<td>$\omega_r$</td>
<td>position relaxation factor</td>
</tr>
<tr>
<td>$\omega_v$</td>
<td>velocity relaxation factor</td>
</tr>
</tbody>
</table>
1 Introduction

1.1 History and description of Brownian motion

Brownian motion was first observed and described by Jan Ingen-Houtz, who was a physiologist, biologist, and chemist, in 1789 (Ingen-Houtz, 1789). His description was based upon the irregular motion of coal dust particles on the surface of alcohol, and the motion was attributed to so-called animalcules. However, Brownian motion is traditionally regarded to be discovered by the botanist Robert Brown in 1827 (Brown, 1828). Brown was studying pollen particles of the plant *Clarkia pulchella* in water under a microscope. He then repeated the experiments with inorganic material and ruled out the commonly held belief that the observed motion was life-related. It is because he extended his experiments to determine the cause of the particle movement, he is accredited with discovering this motion. The founders and notable developers of kinetic theory, namely Maxwell, Boltzmann, and Clausius, never published anything that directly addressed Brownian motion, though some of their ideas permeated into later understandings of Brownian motion (Mazo, 2002). Christian Wiener and Sigmund Exner tried to measure the velocity of a particle undergoing Brownian motion, but were unable to understand the results they obtained (Mazo, 2002). In 1880, Thorvald N. Thiele, a mathematician, became the first person to mathematically describe Brownian motion in a
published paper (Lauritzen, 1999). In 1905, Albert Einstein, a physicist, and in 1906, independently of Albert Einstein, Marian Smoluchowski, another physicist, brought the ideas of Brownian motion to physicists and presented it as a way to confirm the existence of atoms and molecules (Einstein, 1905; Smoluchowski, 1906). In 1906, Jean Perrin used several approaches to determine Avogadro’s number, including direct measurements of the mean square displacement, and applications of Einstein’s equations. In 1908, Paul Langevin showed a link between the different methods used to describe Brownian motion by Einstein and Smoluchowski. Langevin’s main idea was to treat the mean force deterministically and the fluctuating part of the force stochastically (Langevin, 1908). To date, the Langevin equation is the most widely used model to describe the motion of Brownian particles. Perrin became the first to experimentally show Brownian motion conclusively (Perrin, 1909). In 1911, Jean Perrin further showed experimental results that confirmed that Brownian motion is a random process on all length scales and the trajectory is defined by random walk. Figure 1 shows the results from one of the experiments (Perrin, 1916). In this figure, the three particles are of the same radius of 0.53 µm. Each position recorded was taken every 30 seconds and are joined by a straight line segment, and the mesh size of the grid is 3.20 µm.
From these results, it is evident why Brownian movement is hard to observe experimentally. This is because the velocity is related to the time sampling period and difficult to calculate from observed traces using kinematic relations.

Brownian motion occurs as a result of the smaller fluid molecules bouncing off the larger suspended particle. For a gas, Brownian motion is best described and known by the equation for the average displacement of a particle, due to the fact that the velocity of the
particle is infinitely differentiable. The root mean square displacement, $x_{rms}$, in any direction in
a non-moving fluid, after a time $t$, is described by the equation below developed by Einstein
(Einstein, 1905):

$$ x_{rms} = \sqrt{2Dt} \quad (1) $$

where $D$ is the so-called Stokes-Einstein diffusion coefficient and is given by:

$$ D = \frac{k_BT}{b} \quad (2) $$

where $k_B$ is the Boltzmann constant, $T$ is the absolute temperature, and $b$ is the linear drag
coefficient on the particle. The linear drag coefficient in the case of Brownian motion is
generally described by Stokes’ law, i.e., the low Reynolds number flow drag coefficient:

$$ b = 3\pi\nu d \quad (3) $$

where $\nu$ is dynamic viscosity of the fluid, and $d$ is the particle diameter. The Stokes’ drag
coefficient is used even in situations where it is normally not applicable, because if the Reynolds
number is too large then the Brownian motion that is predicted will be negligible compared to
the actual particle motion. Substitution of the equations (2) and (3) into equation (1) yields an
expression for the RMS displacement that shows that the amount of movement of the
suspended particle is dependent on the viscosity of the fluid, its temperature, and the particle
diameter:

$$ x_{rms} = \sqrt{\frac{2k_BT}{3\pi\nu d}} t \quad (4) $$
The fact that the predicted displacement is the root mean squared displacement, suggests there is fluctuation around the mean, which in turn, implies that Brownian motion is a stochastic process. Therefore, it is modeled as such.

1.2 Applications of Brownian Motion

Brownian motion has applications in many fields. Even though it is named after a botanist, this is not the most important field of study. In surveying the main names throughout the history of Brownian motion, physicists played a crucial rule in discovering Brownian motion and developing most of the theory behind Brownian motion. However, it is a phenomenon that is observed and studied by people with different backgrounds. The random nature and the implications of the random nature of Brownian motion have been studied in depth by mathematicians. The effects of Brownian motion are seen in the medical field, most notably in blood flow of humans, and in drug delivery (Lockman et. al., 2002). Therefore, it is also studied by medical professionals. Also, because the main idea behind Brownian motion is the idea of random walk, sometimes the term Brownian motion is used in the discussion and analysis of the random walk of the stock market, and is, therefore, looked at by financial experts. Due to the increased applications of nano and micro-scale technology in objects produced today, the ideas and effects of Brownian motion are becoming more and more important to engineers. The fluid flow in certain systems may approach length scales where attention to the motion of groups of particles may be necessary, even though the length scales may not be small enough to consider each particle of the flow.

One common application where Brownian motion is important is micro-scale filters. One type of micro-scale filter is a filter made by packing a bundle of filters in one area. This type
of filter is different from conventional filters because rather than have the fluid flow through the membrane, causing particles that are too big to be stopped, the particles are filtered out by flowing the fluid parallel to the permeable membrane. This means a membrane is shaped into a pipe and the fluid forced through the pipe, parallel to the membrane. Because there is minimal fluid velocity in the radial direction, particles will move around randomly in the radial direction. Due to the Brownian motion on the particle, the particle will move around and possibly toward the wall (the membrane) and be captured or filtered out from the carrier fluid. As mentioned earlier these cylindrical filters are generally arranged in bundle so many filters are being used simultaneously, thereby increasing the amount of fluid that can be filtered at a single time. Figure 2 shows a picture of a single cylindrical filter where fluid flow is into the figure (Jiang, et al., 2005).
Another application that uses Brownian motion is an H-filter. This filter was first suggested and designed in the 1990’s. These filters take advantage of Brownian motion and use this natural force as the means to filter out a particle (Brody and Yager, 1996). Figure 3 illustrates a schematic of the H-Filter concept (Holl et. al., 1996).
In this figure there are two inlets and two outlets, and due to the very low fluid velocity, the two fluids hardly mix. However, a submicron particle will undergo enough Brownian motion that the particle could diffuse from one fluid’s streamlines to the other and be carried out. These types of filters are of great interest because there are no moving parts and no membranes that need to be cleaned or replaced. However, these filters do have limitations due to the size of the filter, size of the particle looking to be filtered out, the strong dependence of filter efficiency on the particle size, and low input flow rates required. Capture efficiency of two identical fluids in a symmetric geometry is theoretically maximum at 50% because it is equally likely that the particle be filtered or not filtered out simply from symmetry considerations.

Figure 3: H-filter Schematic (Holl et. al., 1996)
Another application where Brownian motion is of interest is in the metal organic vapor phase epitaxy (MOVPE), a type of chemical vapor deposition (CVD), of aluminum nitride (AlN). AlN is a III-V semiconductor that has numerous applications including usage as a light-emitting diode and as surface acoustic wave sensor. It is typically grown using the precursor gases of trimethyl-aluminum (TMAL or (CH$_3$)$_3$Al) and ammonia, NH$_3$. In the MOVPE process, the reactants or the precursor gases, and a carrier gas (e.g. hydrogen), enter a chemical reactor chamber through an inlet showerhead, and then undergo gas-phase reactions above the substrate and gas-surface reaction to deposit AlN epitaxially on a substrate. These chemical reactions occur above the substrate because the substrate is heated. Figure 4 depicts schematically the MOVPE of AlN.
This process is performed in reactors of various geometries and operating conditions. It has been seen in experiments that solid AlN particles form as suspended solids in the gas near the substrate and it is believed that thermophoretic forces keep the suspended particles away from the substrate, causing less AlN growth and waste of precursor gases (Creighton et. al, 2004). Due to the size of the particles formed, Brownian and thermophoretic forces are thought to contribute to the motion of these suspended AlN particles (Creighton et. al. 2007).

1.3 Objectives

The objectives of the present work are as follows:
o Develop a 2-D Brownian motion model based on the Langevin equation

o Validate the Brownian motion model against the analytical solutions to ascertain that the model captures relevant trends accurately

o Extend the model to accommodate:
  - Cunningham slip correction for rarefied flows
  - Thermophoretic forces to address non-isothermal scenarios
  - Lift forces due to inertial effects
  - Any mesh topology (including unstructured) to enable simulation in complex geometries
  - 3-D geometries of arbitrary complexity
  - Unsteady background flow

o Explore the model for the following applications:
  - Cylindrical filter
  - H-filter
  - CVD of AlN

1.4 Organization of Thesis

The remainder of this thesis is divided as follows: Chapter 2 describes the model used to simulate Brownian motion, including the governing equations used, the commercial flow solver that the Brownian motion model was interfaced with, and the numerical algorithms. Chapter 3 presents studies for validation of the Brownian motion model. Chapter 3 also presents the results of the numerical simulations for the afore-mentioned applications and discussion focusing on the results of the simulations. Chapter 4 summarizes the results of the various
simulations and the implications of the research results. Chapter 5 presents thoughts on possible extensions of this work.
2 Mathematical Model and Solution

This chapter presents the mathematical formulation and solution procedure used in the Brownian particle simulations. The first section describes the equations used to describe the motion of the particles. The next section describes the governing equations used to describe the motion of the carrier fluid. Section 2.3 presents the solution procedure.

2.1 Equations of motion of Brownian particle

These equations used for the present study have been derived with the following assumptions:

- Laminar flow (Re<2000)
- Particles in Stoke’s Regime (Re_p<<1)
- Small particles, such that the particle does not interfere with flow around it
- Small local particle volume fractions, such that particles do not interact with one another
- Particle temperature is the same as the local fluid temperature

The equation governing the trajectory of the particle is given below in vectorial format (Wu, Kuznetsov, and Jasper, 2010):

\[
\frac{dr_p}{dt} = u_p
\]  

(5)
where in the above equations \( \mathbf{r}_p \) represents the position of the particle, and \( \mathbf{u}_p \) represents the velocity of the particle. The equation of motion of the particle is the classical Langevin equation and is written as:

\[
m_p \frac{d\mathbf{u}_p}{dt} = \mathbf{F}_B + \mathbf{F}_L + \mathbf{F}_F + \mathbf{F}_T
\]

(6)

where \( m_p \) is the mass of the particle. \( \mathbf{F}_B \) is the Brownian force. \( \mathbf{F}_L \) is the Saffman lift force. \( \mathbf{F}_F \) is the drag force on the particle. \( \mathbf{F}_T \) is the thermophoretic force. Each of these forces are described in greater detail in the equations to follow. The Brownian force is written as:

(Uhlenbeck, 1930; Chandrasekhar, 1943; Gupta, 1985)

\[
\mathbf{F}_B = m_p \mathbf{b}(t)
\]

(7)

where the vector \( \mathbf{b}(t) \) is described by the Cartesian components \( b_i(t) \) as follows:

\[
b_i(t) = G_i \sqrt{\frac{216 \mu k_B T}{\Delta t \rho_p d_p^3 c}}
\]

(8)

where \( G_i \) is a Gaussian random number with zero mean and unit variance. \( \mu \) is the dynamic viscosity of the carrier fluid at the particle location. \( k_B \) is the Boltzmann constant. \( T \) is the absolute temperature of the carrier fluid at the particle location. \( \Delta t \) is the time step over which the motion occurs. \( \rho_p \) and \( d_p \) are the density of the particle and the diameter of the particle, respectively. \( c \) is the Cunningham slip correction factor. The Cunningham correction factor is used to account for non-continuum effects when calculating drag on small particles, and for gases is written as (Crowe, 1998):

\[
c = 1 + Kn \times \{1.227 + 0.42e^{-0.85/Kn}\}
\]

(9)
where $Kn$ is the Knudsen number and is given as:

$$Kn = \frac{2\lambda}{d_p}$$  \hspace{1cm} (10)

where $\lambda$ is the mean free path of the particles and is computed as:

$$\lambda = \frac{1}{\sqrt{2\pi} \sigma^2 n_c}$$  \hspace{1cm} (11)

where $\sigma$ is the molecular diameter of the fluid and $n_c$ is the number of molecules per unit volume. For gases $n_c$ is computed by the ideal gas law:

$$n_c = \frac{p}{RT} N_A$$  \hspace{1cm} (12)

where $R$ is the universal gas constant, $p$ is the local thermodynamic pressure, and $N_A$ is Avogadro’s number.

The Saffman lift force is a force that originates from particle rotation in the flow field (Saffman, 1965). The Saffman lift force is due to a pressure distribution caused on particle surface due to a local velocity gradient. This force causes the particle to experience a lift force perpendicular to the direction of flow of the carrier fluid. The Saffman lift force is given by the following relation (Crowe, 1998):

$$F_L = C_{Saff} d_p^2 \sqrt{\mu \rho_c |\omega_c|^2} \left[ (u_c - u_p) \times \omega_c \right]$$  \hspace{1cm} (13)

where $C_{Saff}$ is the Saffman lift coefficient. A value of 1.61 for $C_{Saff}$ is used. $u_c$ is the velocity of the carrier fluid at the location of the particle. $\rho_c$ is density of the fluid at the particle location. $\omega_c$ is the local vorticity of the carrier fluid at the particle location and is given by the formula:
\omega_c = \nabla \times \mathbf{u}_c \quad (14)

The drag force is given by Stokes’ Law, with the Cunningham correction factor added due to the small particle size and is written as:

\[ F_F = \frac{3\pi \mu d_p}{C_c} (\mathbf{u}_c - \mathbf{u}_p) \quad (15) \]

The thermophoretic force is due to uneven Brownian forces on a particle. This force is caused by a temperature gradient near the particle. This temperature gradient implies that the air molecules around the particles have different kinetic energies, and therefore, different molecule velocities, with higher temperature having a higher kinetic energy and velocity. Because of the random collisions with the particle, this imbalance of molecular velocity causes a force which makes the particle move from higher temperature regions to colder temperature regions. The thermophoretic force on a particle is written as: (Crowe, 1998)

\[ F_T = -6\pi \mu^2 \rho d_p C_s \frac{1}{1 + 3C_m Kn} \frac{k_c/k_d + C_t Kn}{1 + 2k_c/k_d + 2C_t Kn} \frac{\nabla T}{T} \quad (16) \]

where \( C_s \) is the temperature slip constant. \( C_t \) and \( C_m \) are the thermal and momentum accommodation coefficients, respectively. \( k_c \) and \( k_d \) are the thermal conductivity of the fluid and the particle. In this study it is assumed that \( C_t \) is 1.17 and \( C_m \) is 1.14 while \( k_c \) and \( k_d \) are the same value. \( \nabla T \) is the local temperature gradient.

Another equation is needed to validate the assumptions used throughout the simulation. These are the assumptions of a laminar flow of the carrier fluid and that particles are operating in Stokes’ regime. The first equation needed is the Reynolds number for the geometry, \( Re_L \), and is given by the equation:
\[ Re_L = \frac{\rho_c |u_c| L}{\mu} \]  \hspace{1cm} (17)

where \( L \) is the hydraulic diameter of the geometry. \( Re_L \) is needed to check the assumption that the carrier fluid must be operating in the laminar regime and the assumption is reasonable if \( Re_L < 2000 \).

To determine if the particle is operating in Stokes’ regime, the Reynolds number based on the particle size, \( Re_p \), must be much smaller than unity (\( Re_p << 1 \)). \( Re_p \) is given by the equation:

\[ Re_p = \frac{\rho_c |u_c - u_p| d}{\mu} \]  \hspace{1cm} (18)

However, this requires knowing the particle velocity, which is not a known value a priori; therefore a worst-case scenario calculation is done. For the worst-case scenario calculation, the velocity of the particles is assumed to be 0 and the carrier fluid velocity is assumed to be the maximum velocity seen in the geometry. This corresponds to the worst-case scenario for any flow with significant flow speeds because the forces on the particle are generally not significant enough to make the particle travel in the opposite direction of the bulk flow.

2.2 Governing Equations for Carrier Fluid

The governing equations for the carrier are that of the conservation equations of mass (continuity equation), momentum (Navier-Stokes equations), and energy (enthalpy). These equations are presented below in compact vectorial form (Munson et. al., 2009).

Mass:

\[ \frac{\partial}{\partial t} \rho_c + \nabla \cdot \rho_c \mathbf{u}_c = 0 \]  \hspace{1cm} (19)
Momentum: \[ \frac{\partial \rho u_c}{\partial t} + \nabla \cdot \rho_c u_c u_c = -\nabla p + \nabla \cdot \boldsymbol{\tau} \] \hspace{1cm} (20)

Energy: \[ \frac{\partial \rho_c h}{\partial t} + \nabla \cdot \rho_c u_c h = -\nabla \cdot \boldsymbol{q} + \nabla u + \frac{dp}{dt} \] \hspace{1cm} (21)

where \( t \) is the time. \( \boldsymbol{\tau} \) is the shear stress, \( p \) is the pressure, and \( h \) is the enthalpy. The auxiliary equations in the above equations may be written as:

Shear Stress: \[ \boldsymbol{\tau} = \mu(\nabla u_c + \nabla u_c^T) - \frac{2}{3} \mu(\nabla \cdot u_c) I \] \hspace{1cm} (22)

Diffusive Energy Flux: \[ \boldsymbol{q} = k_c \nabla T \] \hspace{1cm} (23)

where \( k_c \) is the thermal conductivity of the fluid. The afore-mentioned conservation equations were discretized using the finite-volume procedure and solved using the commercial CFD (computation fluid dynamics) code CFD-ACE+\textsuperscript{®}, to be discussed later.

### 2.3 Solution Procedure

Modeling Brownian motion required 3 major steps: solving the governing equations for the carrier fluid flow, solving the governing equations for Brownian motion, and post-processing the results. For a steady background flow the process goes straight from one step to the next. However, if the background flow is unsteady, the carrier fluid equations and particle equations for that time step are solved, and then the simulation is stepped forward in time. The new carrier fluid flow field and resulting particle motion at each time step is processed. Figure 5 shows a flow chart, depicting the overall procedure used.
Figure 5: Flow Chart Depicting Overall Model

The first major step, namely, solving the carrier fluid flow, involves 4 main steps: creation of the geometry and mesh generation, setting up the boundary conditions and material properties, as well as solution parameters (convergence criteria, iterations, relaxation factors, etc), solving the equations, and exporting grid connectivity information and the background flow solution information at each cell center (fluid velocity, temperature, viscosity, and density). The information exported will be discussed in detail in section 2.3.1. Figure 6 shows a flow chart outlining the main steps in the carrier fluid flow solution.

Figure 6: Flow Chart Depicting Carrier Fluid Solution

The second major step, namely, solving the Brownian particle equations, involves 4 main steps, as well: importing the data generated by the carrier fluid flow solutions, pre-iterative processing, solving the particle equations including resolving forces and tracking the
particle, and exporting data as necessary. The Brownian particle solver steps are explained in further detail in section 2.3.2. Figure 7 shows a flow chart outlining the main steps of the particle solver as detailed above.

![Flow Chart Depicting Brownian Particle Solver](image)

Figure 7: Flow Chart Depicting Brownian Particle Solver

Details of the solution strategy and each step are presented below in a generalized way that applies to all the simulations. Specifics of every simulation (boundary conditions, forces acting on Brownian particle) are mentioned in the appropriate sections in Chapter 3.

### 2.3.1 Imported Data for Brownian Motion Solver

As shown in the Brownian Particle solver flow chart (Figure 7), the first step in solving Brownian motion is coupling the particle solver to the carrier fluid. This is done by reading the so-called “connectivity” data and carrier fluid flow data. This imported data is the same data as exported from the carrier fluid solver. It is through this data and the export/import process that the carrier fluid solver is fully coupled to the Brownian particle solver.

The “connectivity” data includes numerous data sets and the information in these files is what allows for the particle solver to be used on completely arbitrary geometries and unstructured meshes. This “connectivity” information is needed because each cell used in the
domain is randomly numbered, each cell face has a global number, and each boundary face has its own global number. The information in the “connectivity” data set includes:

- Number of cells
- Number of nodes
- Number of cell faces
- Number of boundary faces
- Number of faces for each cell
- Number of vertices for each cell
- Number of vertices for each cell face
- Cell center locations
- Cell volumes
- Location of each cell face
- Area of each face
- Surface normal at cell face
- Vertex locations
- Cell to cell face links
- Cell to vertices links
- Cell to cell links
- Cell face to cell links
- Cell face to vertices links
- Cell face to boundary face
- Boundary face type
The other imported information is the carrier fluid flow information. This information is needed to calculate the forces on the particle as it moves in the carrier fluid. The imported information in the carrier fluid flow data set is:

- Fluid temperature at cell center
- Fluid density at cell center
- Fluid viscosity at cell center
- Fluid velocity at cell center

2.3.2 Brownian Particle Equation Solver

Now that all the information that is needed to fully couple the Brownian particle solver to the carrier fluid solver the second main step of solving the Brownian motion is setting all needed pre-solver information. This is where features of the Brownian particle solver are set. Seed values for the Gaussian random generator are set. Also specified at this time is which forces are active for the simulation. The start time and stop time of the simulation is set, as well as the time step to be used by the particle solver. This time setting step is vital for time marching simulations because the particle might not be active in the domain at the beginning of the simulation. Also, the time step size might be different than the carrier fluid time step and this allows for the particle tracking to be done with a smaller time step. A feature of this code is that an infinite numbers of particles can be tracked and simulated in the domain in one run; so multiple runs can be done concurrently. The last thing accomplished in the equation solver setup step is specifying the initial conditions of each particle. Each particle’s starting location, starting velocity, density, and diameter are specified. Typically a particle’s starting cell is
specified and the particle is then assigned the cell’s center location with a starting velocity equal to that of the carrier fluid velocity at that cell center.

With pre-processing done, the Brownian motion solver is called. The first step logically is to see if any particles are still active in the domain or if the final time has been reached. If there are no particles active then the program does not need to step any further in time, and clearly if the specified final time is already hit, then there is no need to solve the equations. The second step is to change the active particle in memory and check to see if that particle is still in the domain, if the particle is not in the domain a new particle is put in active memory until a particle still in the domain is found or until all particles have been looked at for that time step. For a particle still active in the domain the ordinary differential equation (ODE) solver is called. The details of this step are presented in a later section. Logically, it is now checked to see if all particles have been looked at for this time step. Once all particles have been analyzed any information that is wanted from the current time step is exported and the process repeats. Figure 8 shows a flow chart outlining the basic steps used to solve the particle equations, including where it fits in compared to Figure 7.
The ODE solver solves the Brownian particle equations developed earlier for the active particle using a time-implicit, backward Euler method. The equations for particle motion trajectory and equation of motion in time-discretized form are written as:

\[ X_{W}(\mathbf{u}_p^{\text{new}}, \mathbf{r}_p^{\text{new}}) = m_p \left( \frac{\mathbf{u}_p^{\text{new}} - \mathbf{u}_p^{\text{old}}}{\Delta t} \right) - F_{\text{new}}(\mathbf{u}_p^{\text{new}}, \mathbf{r}_p^{\text{new}}) \]  

(24)

\[ X_r(\mathbf{u}_p^{\text{new}}, \mathbf{r}_p^{\text{new}}) = \frac{\mathbf{r}_p^{\text{new}} - \mathbf{r}_p^{\text{old}}}{\Delta t} - \mathbf{u}_p^{\text{new}} \]  

(25)
where the superscripts new and old refer to the current time step and previous time step. 

\( F^{new} \) is a generic force function representative of whichever forces are activated for the current simulation: drag, Brownian, thermophoretic, or lift forces, or any combination thereof. As shown by the formulas above and equations derived earlier, the forces acting on the particle are dependent on both the particle’s current position and velocity.

Equations (24) and (25) represent a couple set of non-linear ODEs and are solved by using an iterative Newton-Raphson method. The equations for particle motion trajectory and equation of motion are therefore as given:

\[
\begin{align*}
\mathbf{u}_p^{k+1} &= \mathbf{u}_p^k - J^{-1} X_u(\mathbf{u}_p^p, \mathbf{r}_p^p) \omega_v \\
\mathbf{r}_p^{k+1} &= \mathbf{r}_p^k - J^{-1} X_r(\mathbf{u}_p^p, \mathbf{r}_p^k) \omega_r
\end{align*}
\]

(26)  

(27)

where \( k \) refers to the iteration number. When the iteration number is 1, the old time step value is used. When it is finally converged the \( k+1 \) value is the new value. \( J \) is the calculated Jacobian based on the previous iteration velocity and position and is described in more detail later. \( \omega_v \) and \( \omega_r \) are relaxation factors for the velocity and position and are typically 0.8 and 1, respectively. The residuals of equations (26) and (27) are as all the terms transposed to one side of the equations. For convergence testing, individual residuals are squared and added together to calculate the overall residual value (i.e. \( l^2 \) norm). Typically, a maximum overall residual of \( 1e-7 \) was required before the problem is considered to have been converged.

To solve the equations (26) and (27), the ODE solver first calculates the Gaussian random numbers based upon the earlier selected seed numbers that are needed for the Brownian forces and these remain constant throughout the iterative process. The flow velocity
and flow properties are calculated based upon the current particle location, and this is the first step of the iterative process. The flow information at the particle location is interpolated from the known values at cell centers by using a distance averaged weighting scheme of the particle’s current cell and surrounding cells. With this information, the active forces that act upon the particles are calculated. The drag and Brownian forces are straightforward calculations but the Saffman lift force and thermophoretic forces are not trivial due to the required calculations of the velocity and temperature gradient. To calculate these values the divergence theorem is used which can be expressed as:

\[
\int_{V} \nabla \cdot \mathbf{q} dV = \int_{S} \mathbf{q} \cdot n dA
\]  

(28)

where \( \mathbf{q} \) is an arbitrary vector, \( V \) is the volume of particle’s current cell and \( S \) is the surface bounding that volume. By considering \( \mathbf{q} = \phi \mathbf{i}, \phi \mathbf{j}, \text{ or } \phi \mathbf{k} \), it can be shown that:

\[
\nabla \phi = \frac{1}{V} \sum_{faces} n_{f} \phi_{f} A_{f}
\]  

(29)

where \( n_{f} \) is the unit face normal, \( \phi_{f} \) refers to the face value of \( \phi \), and \( A_{f} \) is the area of the face. The face values are calculated from cell center values using weighted interpolation. Equation (29) applies to calculation of any scalar \( \phi \). In the current context, this equation was used to compute gradient of temperature for thermophoretic force calculation, and gradient of the individual velocity components for vorticity calculation.

Once the calculation of all forces active in the simulation are accomplished, the residual values based upon the governing equations are calculated as previously shown. The Jacobian of the governing equations is then calculated. The Jacobian equations were analytically derived and then calculated at each iteration. The velocity and position correction values are then
solved by Gaussian elimination by using the residual values and Jacobian. With the current position and position correction values known, a particle tracking algorithm was called to see how the position correction value should be applied. This tracking algorithm updated the particle velocity (if needed) and which cell the particle is in after applying the position correction. Details of this procedure are presented in section 2.3.2.1. With this information the particle velocity and position was updated using the correction values. The residual values that were calculated are now checked to see if the total residual value is lower than the maximum residual value accepted. If it is below the maximum allowed value then the solution is said to have converged and the solver exits, if it is not converged, the process goes back to the calculating of the flow information step and repeats until it converges. Figure 9 shows a flow chart that illustrates the main steps of the process that is used by the developed ODE solver.
Calculate needed Gaussian random numbers

Get flow information based upon current particle location

Calculate forces acting on particle

Calculate residuals from both position and velocity equations

Calculate current Jacobian values

Solve for the velocity and position correction

Call particle tracking algorithm

Update velocity and position using correction values

Check residual for convergence

No

Yes

Exit ODE solver

Figure 9: Flow Chart Depicting ODE Solver
2.3.2.1 Particle Tracking

The algorithm used for particle tracing is a modified version of ray tracing as developed for surface-to-surface radiation transport by Mazumder (Mazumder, 2006). The first step in this algorithm is to gather the particle information needed for the rest of the algorithm, this includes the particle starting location, \( \mathbf{r}_p^{\text{start}} \), the direction and distance a particle will travel in one time step, \( \mathbf{d} \), and the particles starting cell, \( \text{cell}^o \). For the first time through this algorithm, the distance to travel is simply the particle’s velocity multiplied by the time step as given by the formula:

\[
\Delta t * u_{p}^{\text{new}} = \mathbf{d}
\]

Knowing the starting location, the direction of travel, and the current cell, each face of the cell is checked to see which face the particle could interact with and the intersection point, \( \mathbf{r}_p^{\text{intersect}} \), is calculated. Details of how this is accomplished are provided further on. The distance between the intersection point and the particle, \( l \), is calculated as given:

\[
\mathbf{r}_p^{\text{start}} - \mathbf{r}_p^{\text{intersect}} = l
\]

If the distance the particle travels is less than the distance between its starting location and intersection point, \( ||\mathbf{d}|| < l \), then the particle remains in its current cell and the particle’s final position is given by:

\[
\mathbf{r}_p^{\text{start}} + \mathbf{d} = \mathbf{r}_p^{\text{final}}
\]

Therefore, the final location of the particle tracker is found and the algorithm is complete. However if the distance the particle travels is more than the distance between its starting location and intersection point, i.e. \( ||\mathbf{d}|| \geq l \), then the particle interacts with the cell face. At
this point it is important to know what kind of face the cell face is and depending on what kind of face the cell face is different actions will be taken. If the cell face is a boundary face, the type of boundary face is now needed. If the boundary face is actually material face, (indicating that face is the interaction between two different fluids) then the face is treated as a normal cell to cell face, which is explained in detail later. If the boundary face is an outlet then the particle escapes through the outlet and is removed from the active domain. If the boundary face is any of the other types of boundary faces seen in these simulations, symmetry, inlet or a wall, the face was typically treated the same. These types of faces were handled as polished specular walls, and therefore, the particle would symmetrically reflect off the wall. When a reflection occurred, the particle’s distance to travel direction was modified to reflect the change in direction. The distance travel magnitude was also adjusted to reflect the amount the particle has moved and the particle was moved to the intersection point and the process was restarted with this updated information. The equations given show the changes made to the starting values for this situation:

\[ r_p^{start} = r_p^{intersect} \]  \hspace{1cm} (33)

\[ ||d|| = ||d|| - l \] \hspace{1cm} (34)

This concludes all possible boundary faces that the particle interacts with. The other case, and most common case, is that the cell face the particle interacts with is a cell to cell interface. In this case the particle’s location is moved to the intersection point. The distance to travel magnitude is adjusted because the particle has moved. The cell in which the particle is located is updated to the new cell it is entering. The process is then restarted with the updated values to \( r_p^{start}, d, cell^p \). The process continues until the particle escapes out of a boundary
outlet, or if the distance the particle travels keeps it contained in its starting cell. Figure 10 shows a simple schematic of a particle which crosses over a cell to cell interface, this particle originates in $cell^0$ with position $r^\text{start}_{p}$ and distance to travel $d$ and intersects at the point $r^\text{intersect}_{p}$ using the process as described earlier. This schematic was developed to show physically the variables used in this algorithm.

Figure 10: Particle Tracing Schematic
Figure 11 shows a flow chart describing the major components of the algorithm that has been developed.

In the flow chart as shown if there is not an arrow from a step to something else then this where the algorithm logically ends, and the program returns to the ODE solver as shown in the ODE solver flow chart, Figure 9. In this flow chart, there is one step that needs to be
explained in further details, and this is step of deciding whether a particle’s travel intersects with a cell face.

This intersection calculation uses the following algorithm developed by Mazumder and Kersch to determine if the particle’s travel causes it to intersect with a cell and if it does where it intersects (Mazumder and Kersch, 2000). The algorithm described here in detail is for the 2-D intersection method, but scales to 3-D. This algorithm requires knowing particle starting location, \( r_{p_{\text{start}}} \), particle traveling direction, \( d \), the face location \( A \), and vector describing the face \( \overrightarrow{AB} \). The face location \( A \) is the location of one of the vertices of the face, while \( B \) is the location of the other vertex of the face. Another piece of information is needed, as a reference point for calculations and while any point could be used for simplicity the origin, \( O \), is used. Using vector geometry, various necessary vectors with the location of the intersection point, cell face, and particle starting location as they relate to the origin can be developed and are symbolized as \( \overrightarrow{Or_{p_{\text{intersect}}} A} \), \( \overrightarrow{OA} \), and \( \overrightarrow{Or_{p_{\text{start}}} A} \), respectively. Also the vector \( \overrightarrow{r_{p_{\text{intersect}}}} A \) is used to describe the position of face location relative to the intersection point. Using these equations, a relationship between the intersection point and the wall can be developed and explicitly expressed as:

\[
\overrightarrow{Or_{p_{\text{intersect}}} A} = \overrightarrow{OA} - \overrightarrow{r_{p_{\text{intersect}}}} A
\]  

(35)

This can be re-written in an alternate form as:

\[
\overrightarrow{Or_{p_{\text{intersect}}} A} = \overrightarrow{OA} + \beta \overrightarrow{AB}
\]  

(36)
where $\beta$ is a constant value and if $\beta$ is between 0 and 1 then the particle could intersect with the face as long as another condition is met. A vector relationship can be developed relating the intersection point to the particle starting location and is written as:

$$\overrightarrow{r_p^{\text{intersect}}} = \overrightarrow{r_p^{\text{start}}} + l \cdot d$$  \hspace{1cm} (37)$$

where $l$ is a constant value and if $l$ is positive and the previous prescribed condition is met, then the particle could intersect with the cell face. By combining equations (36) and (37) and substituting values in and re-arranging, a simple expression is developed and is expressed by:

$$\beta \overrightarrow{AB} = \overrightarrow{r_p^{\text{start}}}A + l \cdot d$$  \hspace{1cm} (38)$$

This simple expression represents a 2-D vector equation and can be used to solve for the two unknowns $\beta$ and $l$. As mentioned earlier, the values of $\beta$ and $l$ can be analyzed to see if the particle’s travel direction causes it to intersect with the cell face. The necessary and sufficient conditions for the particle to potentially interact with the face is that $0 \leq \beta \leq 1$ and $l > 0$. Figure 12 shows a schematic of the system.
2.4 Carrier Fluid Solver Details

In the first major step as detailed by Figure 5, solving the carrier fluid flow, the first step as shown in Figure 6 was to create the geometry and generate the mesh. The geometry file and the resulting mesh were created using the program CFD-GEOM for both 2-D and 3-D simulations. Figure 13 below is a screen shot of CFD-GEOM.
The meshes created were created by an automatic mesh generator and as described in the Brownian particle solver, the meshes were assumed to be unstructured. The geometry and mesh information was saved and the data was transferred into another program called CFD-ACE-GUI, via a proprietary file format, known as a .DTF file format. Figure 14 below is a screen shot of CFD-ACE-GUI, showing the main display screen of the program.
CFD-ACE-GUI is where the second major step of solving the carrier fluid flow is done, the setting up of the model parameters. The purpose of CFD-ACE-GUI is to set the boundary conditions, initial conditions, material properties, and solver parameters. Some example conditions set by the user in CFD-ACE-GUI include fluid temperature, fluid viscosity, and inlet flow rate.

The next major step of solving the carrier fluid flow is the actual solving of the governing equations. The solver employs an iterative technique to attain convergence based upon the solution techniques chosen by the user. The residuals of each of the governing equations are
tracked as they progress towards the desired convergence levels. Figure 15 shows a typical normalized plot of the residuals.

![Residual Plot](image)

**Figure 15: CFD-ACE Sample Residual Plot**

CFD-ACE generated all the carrier fluid outputs that are seen in this report. The outputs of CFD-ACE are saved in either the original .DTF file or new .DTF files for each time step if a time-marching simulation is done.

The final step in solving the carrier fluid flow is post-processing. This is done primarily in CFD-VIEW, the CFD-ACE post-processing program. This was used to see how different flow variables such as temperature or velocity varied over the geometry. Figure 16 is a snapshot of
CFD-VIEW showing fluid flow. For time marching cases the separate .DTF can be linked together in CFD-VIEW to generate an animation video of the entire sequence of events.

Figure 16: CFD-VIEW Sample Screen Shot
3 Results and Discussions

This section contains information pertaining to the simulations performed. The model developed is first validated for both 2-D and 3-D simulations against analytical results. It is then demonstrated for different applications. The first application investigated is an H-filter application. The second application studied is a cylindrical filter application. The final application for which the model is explored is the motion of AlN particles in chemical vapor deposition. For this particular application, both horizontal and vertical reactors are studied for both steady and unsteady flows.

3.1 Validation of Brownian Motion Model

The first set of simulations was performed to ascertain that the results of the code matched analytical results. This was done by creating test geometries and boundary conditions where the resulting flow fields are simple.

3.1.1 Validation in 2-D

For the validation of the Brownian motion model, the first test considered was that of flow between two parallel plates. The carrier fluid properties used are a density of 1.12 kg/m$^3$ and fluid viscosity of 1.68x10$^{-5}$ kg/m-s. The other inputs to the system include an inlet to outlet pressure difference of 0.02 Pa, a particle density of 1000 kg/m$^3$. The gap between the plates is 1
cm (y-direction) and the length of the plates is 6 cm (x-direction). The resulting carrier fluid flow is immaterial for the validation cases because we are primarily concerned with how the particle moves in the y-direction where the fluid velocity is negligible. The x-direction flow is included to prove that the particle is properly affected by the drag caused by the fluid flow and to show that compared to the bulk motion of the fluid that Brownian motion is significantly smaller. The maximum x-direction velocity of the carrier fluid is approximately 0.6 m/s. The Brownian motion simulation step size used for the ODE solver was 5 µs, and a particle diameter used is 0.05 µm. $Re_L$, based on the hydrodynamic diameter of the geometry, is approximately 400. Therefore, the carrier flow is laminar. $Re_p$, using a stationary particle with a diameter of 0.05 µm, with the highest flow velocity is approximately 0.02 m/s. This corresponds to a worst-case scenario, because the particle is inserted at the fluid velocity and follows the carrier fluid, with deviations caused only by Brownian motion. Therefore, $Re_p$ is generally much smaller and Stokes’ law for calculating the drag force is appropriate.

The first case investigated in the parallel plate geometry is a single particle moving in the flow subject only to drag and Brownian forces. The particle is started in the middle of the inlet of the channel with starting velocity equal to the fluid velocity. Figure 17 shows a trace of a 0.05 µm diameter particle in the parallel plate geometry.
Figure 17 shows that the particle is clearly moving in the direction of the bulk fluid flow (in the positive x-direction), but also randomly moving around in both the x and y-directions. This particle is followed for approximately 0.01 s and it is seen that the particle travels the expected approximate distance of 6 mm during this time period, showing that the drag force on the particle is implemented correctly. During this simulation, both the Brownian forces and the velocity of the particle in both the x and y-direction were recorded for each time step. Figure 18 shows the velocities of the particle associated with the particle trajectory shown in Figure 17.
Figure 18: Sample 0.05 μm Diameter Particle’s X and Y-Velocities for a Time Duration of 0.01 s in a 2-D Steady Flow with a Reynolds Number of 400

From Figure 18 it can be seen that the effect of the Brownian forces, and the resulting particle velocity, is relatively small compared to the effects of the fluid drag. Figure 19 shows the Brownian force per unit particle mass (particle acceleration) acting on the particle causing the random velocity in Figure 18, and therefore, the random motion that is observed in Figure 17.
Figure 19 highlights that the random forces are independent of each other, as desired. Also, by inspection, these forces per unit particle mass clearly appear to have zero mean and fluctuate consistently but randomly around 0 N/kg. Both x and y-direction forces also appear to have approximately the same maximum positive and maximum negative forces, implying the force is equally random in all directions.

These simulations were then repeated with 1000 particles, each with the exact same start location, same particle properties, and same fluid conditions. For this simulation the x and y-location of each particle for each time step is recorded. Also recorded at each time step for
each direction are the maximum and minimum values, as well as the calculated average value and standard deviation of each particle’s velocity and position. Figure 20 show two plots, where the bottom graph is a magnified version of the top graph. In this figure, the average x location at each time step is shown, as well as the maximum, minimum particle locations, and the standard deviation of the x location.

Figure 20: 0.05 µm Diameter Particle Trajectory in the X-Direction for a Time Duration of 0.01 s in a Flow with a Reynolds Number of 400 for a 2-D Steady Simulation with 1000 Particles
From analyzing the top graph there is no distinguishable difference between the average particle location and the minimum and maximum locations. It is only with the bottom graph that these values can be seen. It is clear from this plot that the particles undergo Brownian motion, and that the effects of the Brownian force is much smaller than the drag force, as seen by the plots earlier. It is for this reason, Brownian effects are generally not investigated in the direction of the fluid flow, if there is fluid flow, and the Brownian effects are observed in a direction where there is minimal fluid flow. In these simulations, the direction of minimal fluid flow is the y-direction, and below in Figure 21, the average y location is shown as well as the maximum and minimum locations and the standard deviation which, because of no y-direction flow, is also the root mean squared (RMS) distance.
Because Figure 21 is in the direction where there is minimal fluid velocity, it shows the results normally associated with Brownian motion. It is clear that particles tend to move away from the average location and that the RMS distance grows as time grows.

To study this in greater detail, a new and simpler geometry was created. The geometry created is a 10 cm, square cavity. The carrier fluid inside the square was assumed to be static, and therefore, without Brownian forces, the particle would not move. However, once the
particle starts moving there are still drag forces on the particle. The particle still undergoes drag forces because the particle moves due to Brownian forces and its movement is then impeded by the drag force from the stationary fluid. The geometry size is such that it is much larger than the particle will travel in 8 s by Brownian forces alone, and therefore, there will be no wall effects felt by the particle. The fluid properties needed for the simulation are a viscosity of $1.1614 \times 10^{-5}$ kg/s-m and a temperature of 298 K. 5000 particles each with a diameter of 0.05 µm are used for this simulation. Since there is no flow in the geometry there is no $Re_L$. $Re_p$ is going to be very, very small for the particle and just based upon the Brownian velocity. Since the motion of the particles is isotropic, the remaining validation tests are only shown in the y-direction for brevity and either the x or y-direction data could have been used because of the setup used.

The first set of simulations conducted in this geometry was to determine if there are any time discretization errors for the Brownian motion solver. Since the model used in the numerical simulations is derived from kinetic theory, the numerical results should match the predictions of kinetic theory exactly. Therefore the calculated RMS values are compared to the analytical equation (4), which was originally presented in section 1.1 on page 4. Figure 22 shows the effect of time step size by comparing numerical results to the calculated analytical results of equation (4) where not every data point recorded is presented for clarity reasons.
Figure 22: 0.05 µm Diameter Particle RMS Distance in the Y-Direction for a Time Duration of 8 s in a Stagnant Flow for a 2-D Steady Simulation Based on 5000 Particles

By inspection Figure 22 shows that the simulated data matches the analytical data very closely, irrespective of the time step size. It can be seen that the predicted values are seen to go slightly above and below the expected values with both small and large time steps. Therefore one can conclude that time steps of up to 1 s can be used and not affect the accuracy of the solutions. It is clear that the simulated responses closely match the analytical results.

With the time-step varying test showing little or no time discretization errors, other validation tests were undertaken to demonstrate how each parameter of the Brownian motion
model affects the calculated RMS values, as well as validate the results obtained from these tests against analytic results. This involved testing the inputs to the Brownian solver that modify the Brownian force; these inputs are the particle diameter, fluid temperature, and fluid viscosity. These properties were changed independently and their effects were studied. This is an approximation because in a real fluid a change in temperature would typically result in a change in viscosity and density. In this case, for clarity of understanding these effects, each property was kept independent. For these 1000 particle simulations, the time step used was 100 µs. Figure 23 shows the results of varying the diameter from 0.01 µm to 0.05 µm with constant fluid viscosity of $1.68 \times 10^{-5}$ kg/m·s and a fluid temperature of 298 K, with comparisons to the analytical results as predicted by equation (4).
This shows that as a particle size is increased, the particle will undergo less Brownian motion. This makes sense because the mathematical models of Brownian motion and drag stipulate that the Brownian force increases proportionately with $\sqrt{d}$ while the drag increases proportionately with the $d$. Therefore, as the particle diameter increases the effect of the Brownian motion compared to the drag force is much less. Figure 23 reflects this phenomenon, showing that the model used correctly predicts the RMS distance for particles of different sizes.
The next test done was to see the effect of varying the fluid temperature. This was done with a 0.05 µm diameter particle, with a 100 µs time step and viscosity of $1.68 \times 10^{-5}$ Kg/m·s. Figure 24 shows the y-direction RMS value with various temperatures in comparison to the predicted values from equation (4).

It shows that as the temperature is increased, a particle of the same size will undergo more Brownian motion. This result is explainable by analyzing the kinetic energy of the fluid. As the absolute temperature of the fluid increases, the kinetic energy of the fluid increases, and as

![Figure 24: Y-Direction RMS with Varying Fluid Temperature for a Time Duration of 20 ms in a Stagnant Flow for a 2-D Steady Simulation Based on 1000 Particles](image-url)
the kinetic energy increases, the fluid molecules travel with a greater velocity. Since the fluid particle weight is constant, but the particle is traveling faster, it will have a greater momentum associated with it. Because of the collisions that occur randomly between the fluid molecules and the Brownian particle, an increased fluid particle momentum will cause an increased Brownian force on the Brownian particle. Apply to the many fluid particles around the Brownian particle it is clear that a greater Brownian force on each side of the particle is possible. This would cause it to have a higher RMS translation value as seen, but at the same time still have the same average location.

The next test done was to study the effect of varying the fluid viscosity. This was done with a 0.05 µm diameter particle, a 100 µs time step, and fluid temperature of 298 K. Figure 25 shows the y-direction RMS values with varying viscosity values and comparison to the calculated analytical values obtained from equation (4).
The predicted and simulated data match, and the results clearly show that as the viscosity of the fluid is increased the Brownian motion is decreased. This result can be explained by the fact that fluid with a higher viscosity imposes more drag on the particle. The viscous drag will tend to slow down the Brownian motion of the particle, and bring it closer to mean velocity of the fluid, which in this case is 0 m/s.

These validation studies show that the numerical model accurately captures the effect of the various parameters in the Brownian motion model, and correctly mimics all relevant physical phenomena associated with Brownian motion.
3.1.2 Validation in 3-D

Although the results of the 2-D validation studies are expected to carry over to 3-D, validations were also conducted in 3-D to ascertain that the 3-D tracking algorithm was functioning correctly.

To validate the 3-D code, a simple test case was used. The test case used was a 0.1 mm cube duct where all the outer faces are solid walls. The fluid flow inside was assumed to be static, and therefore without Brownian forces, the particle would not move. Note: for the 3-D validation tests only x-direction results are presented, but since Brownian motion is isotropic the y and z-directions could be presented.

The operating parameters include a fluid temperature of 300 K, fluid viscosity of $1.846 \times 10^{-5}$ kg/m-s, and fluid density of 1.1614 kg/m$^3$. 1000 particles each with a diameter of 0.05 μm were used. The time varying tests that were done for the 2-D cases were repeated, and Figure 26 shows the results for time steps of 0.2 s and 0.04 s in the x-direction with the results compared to the analytical solution as given by equation (4) where not every data point recorded is presented for clarity reasons.
The results from these simulations are expected, but nevertheless, these simulations were conducted to ascertain that the 3-D tracking algorithm was functioning correctly. Because the model for Brownian motion and drag was not adjusted, the model is not re-validated for varying particle diameter, fluid temperature, and fluid viscosity in 3-D.

### 3.1.3 Validation in 3-D for Unsteady Flow

An extension to the code was needed to couple the Brownian motion solver to unsteady background flow. As with going from 2-D to 3-D, adding this feature did not directly affect the models describing the Brownian force, but rather modified the algorithm of the model. The change to the Brownian model was in the way the Brownian solver interacts with the flow...
solver. Because of this change, the new coupling procedure was validated to make sure the new model used was implemented properly.

The test case used was a 1 m cube duct where there is one inlet and one outlet and all the outer faces are solid walls. The inlet is on the $y$-$z$ plane, centered on the origin. Figure 27 is a 2-D schematic depicting the 3-D geometry.

![Figure 27: 2-D Schematic for 3-D Unsteady Fluid Validation](image)

In this study both the flow solver and Brownian solver use a 0.01 s time step and a particle diameter of 0.05 µm. The Brownian particles were started at the origin at time of 0. The flow starts with an initial velocity only in the $y$-direction of -1 m/s. Once the simulation is
started an x-direction inlet velocity of 0.1 m/s is specified and after one second the inlet turns into outlet with specified velocity of -0.5 m/s. Due to the initial fluid velocity the particle moves in the negative y-direction the entire duration of the simulation. The changing x-direction flow causes the flow to initially start flowing in the positive direction, but ultimately flow in the negative x-direction, causing the particle to exit the outlet. There is no flow specified in the z-direction, therefore the particle should noticeably exhibit Brownian motion in this direction. Figure 28 shows the trajectory of one particle, showing that the particle is correctly tracked over time.
3.2 Applications

From the preceding verification and validation studies it is seen that Brownian motion can cause a particle to move a significant distance. By being aware of this and anticipating this random movement, the effects of this movement can be leveraged to accomplish different tasks. An H-filter application is analyzed and the filter capture efficiency for varying particle diameter is investigated under different flow properties. A cylindrical filter is tested to see the effects of different flow properties on capture efficiency. Finally, the effect of Brownian motion on Brownian particles in both horizontal and vertical chemical vapor deposition reactors under both steady and unsteady flow conditions is investigated to understand how this force affects the motion of particles formed within the reactor.

3.2.1 H-Filter

An H-filter, as described in section 1.2, is a section of micro-channels used as a filter and is of great interest because there are no membranes or similar components that require cleaning or replacement. The H-filter can be used for the continuous extraction, or filtering of interfering particles from a flow. The interfering particles could be blood cells, microorganisms, dust, viruses, etc. Figure 29 shows the schematic of the H-filter used for the present simulations.
These filters are modeled as having polished solid walls, meaning that if a particle collides with the wall, the particle bounces off. For modeling purposes, since no other information is available, it is assumed that the reflection off the wall is completely elastic and specular. The main idea behind an H-filter is that small particles undergo enough Brownian motion to cross over streamlines and exit the other side, while larger particles would stay on the same streamlines and exit with the carrier fluid it came in with. Under this premise, the theoretical maximum filtration efficiency of a particle of any size would be 50%; since probability-wise, the best case scenario is 50% of the particles crossing over to the other fluid.
In the simulations conducted of the H-filter, the geometry used was that of a typical H-filter as shown in Figure 29. This filter model uses the same fluid and boundary conditions for both inlets. For both outlets the same boundary conditions are specified. The inlet and outlet conditions are a fluid speed of 2 μm/s and a gauge pressure of 0, respectively. The fluid properties used are a fluid viscosity of $1.846 \times 10^{-5}$ kg/m·s and a fluid density of 1.1614 kg/m$^3$. The whole setup was assumed to be isothermal at 300 K. The model developed does not require that the inlet fluids are identical or that the wall temperatures to be the same, but if these values are different than the maximum theoretical capture efficiency could be different than 50%.

The $Re_L$ is approximately $10^5$, and therefore, operating in the laminar regime. For particles used in this application the $Re_p$ is approximately $10^6$. With these properties and flow speeds, the two fluids travel through the geometry with minimal mixing occurring between the two fluids. Figure 30 shows velocity vectors of the simulation at the top intersection point.
This shows the fluid does not actively mix when at the top intersection point. From this it can be implied that the bottom intersection point shows the same results, and therefore, the fluid coming in through the right inlet goes out the right outlet and the same applies for the left inlet and outlet. However, at these locations, the carrier fluid changes direction and some vorticity in the fluid is seen.

As mentioned earlier, in this application it is important to see if a particle crosses streamlines, therefore the Saffman lift force is included in calculations. The Saffman lift force is
an inertial effect caused by the viscous flow around the particle and is experienced in the
direction perpendicular to the flow. In this simulation, however, the effect of this lift force
compared to the Brownian force and drag are seen to be marginal and this lift force is
essentially unnoticeable in the results shown. A particle density of 1250 kg/m$^3$ was used for the
simulations. For all the simulations, particles were injected in the center of the top right input
of the filter with a starting velocity equal to the fluid velocity. The time step size used for
particle tracking is 0.1 s. The first particle simulated was a 10 μm diameter particle. As seen by
the validation cases earlier, large particles undergo less Brownian motion and smaller particles
undergo more Brownian motion. A 10 μm diameter particle is large enough that the Brownian
movement on it is minimal, and therefore, should not be filtered out. Figure 31 is an example
trace of a 10 μm diameter particle.
Figure 31: Large Particle (10 μm) Trajectory in an H-Filter

Clearly this particle undergoes very little noticeable Brownian motion, and does not get filtered out. The simulation was then run using 0.1 μm diameter particles. These particles are small enough that they could be filtered out. Two example particle traces are shown below; Figure 32 shows a particle trace of a particle filtered out and Figure 33 shows a particle trace of a particle that was not filtered out.
Figure 32: Small Particle (0.1 μm) Trajectory in an H-filter when Filtered
These trajectories show that, in keeping with the theoretical probability of 50% capture for a small particle, it seems almost random whether the particle will be filtered out or not. It is now established that the simulation mimics the operation of the H-filter. Parametric studies were next performed to investigate how the capture efficiency of the H-filter is dependent on the particle diameter. Figure 34 shows how the capture efficiency varies as the particle diameter varies, these tests were done with a sample size of 500 particles.
Figure 34 shows the expected result, that as a particle size is decreased it will have a greater chance of being captured, because of the increased Brownian motion, and reaches the theoretical maximum of 50%. Given the figures from section 3.1.1 demonstrating how the Brownian motion can be increased or decreased by modifying the fluid properties, viscosity and temperature, the capture efficiency at a certain diameter could be adjusted by changing the fluid properties. Just as was done with the validation studies, these properties were varied independently of each other. Figure 35 shows the effect on capture efficiency of the filter with
particles of different diameters while varying the simulation temperature from 300 K to 600 K for simulations with 500 particles.

Figure 35: H-Filter Capture Efficiency with Varied Fluid Temperature for 500 Particles

This confirms the theoretical results, that by increasing the fluid temperature, and consequently increasing the Brownian motion, the capture efficiency increases. As shown in Figure 35, one particular case predicts a capture efficiency greater than the theoretical maximum of 50% and this is due to the sample size of the particles. If more particles were simulated, the efficiency would be closer to 50%. Figure 36 shows the effect on capture
efficiency of the filter with particles of different diameters while varying the viscosity from $0.923 \times 10^{-5} \text{ kg/m-s}$ to $3.692 \times 10^{-5} \text{ kg/m-s}$ for 500 particles.

Figure 36: H-Filter Capture Efficiency with Varied Fluid Viscosity for 500 Particles

Figure 36 shows the theoretically expected result and that is if the fluid has less viscosity there will be more Brownian motion, and therefore, there will be more particles captured up to the theoretical maximum of 50%. Another way the capture efficiency could be adjusted is to modify the fluid flow speed. If the fluid flow speed is decreased, then the amount of time each particle is actively in the filter would increase, allowing particles to travel further and increasing capture efficiencies (to the maximum of 50%). If the fluid flow is increased, the particle would
travel through faster allowing less time for Brownian motion, therefore, lower the capture efficiency.

3.2.2 Cylindrical Filter Application

The second application explored is a cylindrical filter. As mentioned in section 1.2, the cylindrical filter operates on a different set of principals than the H-filter. The H-filter is designed such that there are no membranes, whereas the cylindrical filter uses a membrane, in fact, the whole filter is constructed out of the filtering membrane. In the simulations, when the carrier fluid flow is determined, the membrane wall is assumed to be a solid wall, and in the Brownian particle tracking algorithm the wall is treated as an outlet. By treating the wall as an outlet, the simulations treat a particle collision with the membrane as the particle being filtered out, thus leaving the active domain. In these simulations, wall collisions were determined based upon the particle’s surface, not the particle center. Figure 37 shows a schematic of the filter with dimensions shown.

![Figure 37: Y-Z Plane of Cylindrical Filter Schematic](image-url)
The inlet and outlet of the filter are on the x-y plane. The input parameters to this simulation include an inlet to outlet pressure difference of 160 Pa. The fluid parameters used are a fluid viscosity of $1.1614 \times 10^{-5}$ kg/m$^3$, fluid density of 1.846 kg/m-s, and isothermal conditions with temperature of 300 K. A constant particle density of 1000 kg/m$^3$ was used. The maximum fluid velocity is around 0.02 m/s. Therefore, $Re_f$ is approximately $5 \times 10^3$ and the flow is considered laminar. $Re_p$ is approximately $10^3$.

Much like what was seen in the 2-D H-filter application; larger particles should undergo less Brownian motion and should not be filtered out. This application is first compared to numerical results presented by Wu as a way to validate the model used against other numerical results. For these simulations, the particle injection locations were randomly chosen across the cross-sectional area of the inlet of the cylindrical filter. This is done because in the physical application the flow field heading into the inlet causes the particles to be anywhere. The injection locations used are at 0 m in the z-direction, with the x and y-locations were bounded to be at 0.99 of the radius. This bounding is done in the work done by Wu because of wall interaction forces not being considered in this simulation. The simulations referenced have these forces turned off, but the injection procedure was maintained. The particle injection velocity is the same as the carrier fluid velocity at the particle location. Figure 38 shows a trajectory of a 0.1 $\mu$m particle depicting the distance traveled through the filter compared to the radial location of the particle, with a random starting location.
Figure 38: 0.1 μm Diameter Particle Trajectory in a 3-D Cylindrical Filter

It shows the particle moves around the radial direction of the cylinder as expected. Figure 39 shows the trajectory of 0.01 μm diameter particle on the y-z plane, with a random starting location.
It also shows the expected result, that a smaller particle will move around much more. This is seen by the sharper movements in the radial direction. The same setup was repeated for particles of varying diameters using 1000 particle sample. Figure 40 shows how the capture efficiency changes as the particle diameter changes from 0.05 μm to 0.6 μm compared to Wu’s simulated data (Wu et.al., 2010).
Figure 40 shows that below 0.02 µm as particle diameter decreases, the capture efficiency increases. It is interesting though, that as the particle diameter increase that the capture efficiency also increases. This can be explained because as particles get bigger, each particle will have less distance to travel before it hits the membrane. Because of the design of this filter, it is possible to get 100% particle capture and this is predicted by extrapolating the data in Figure 40 with a smaller particle diameter. Larger particles than 0.02 µm have a higher predicted capture rate but this is practically unrealistic since a particle this large would cause the filter to be cleaned too frequently and the assumption of particles not changing the carrier fluid
flow breaks down. The results of these simulations are further examined to see where the particle is captured for various particle diameters. Figure 41 shows the capture efficiency of the first 0.1 μm of the filter compared to the total capture efficiency and is compared to the simulated data by Wu.

![Figure 41: 3-D Cylindrical Filter Capture Efficiency in First Ten Percent of Channel (Wu et. al., 2010)](image)

This data shows that the larger diameter particles are much more likely to be captured near the beginning of the filter compared to smaller diameter particles. Figure 42 shows the percentage of particles captured in each 0.1 μm section of the filter out of the total amount of
particles captured by the filter and then compares the results to the simulated data by Wu. It shows that larger particles are more likely to be captured in the first 0.1 μm compared to smaller particles, as shown in Figure 41, and then as the particle travels further downstream these larger particles are very unlikely to be captured. Smaller particles are shown to be more likely captured in the first 0.1 μm compared to the other sections, but the percent captured by the later portions of the filter is noticeably higher than for the larger particles.

Figure 42: 3-D Cylindrical Filter Capture in Each Section (Wu et. al., 2010)
It was seen that for when doing these simulations that the 1000 particles had a noticeable statistical variation. This was analyzed in detail for the case of a 0.1 μm diameter particle, and 10 simulations with the exact same inputs were done. For each simulation ran, the percent captured in each 0.1 μm is calculated by recording the number of particles captured in the section divided by the total number of particles inserted was recorded. These values were then analyzed by recording the maximum and minimum percent captured values found in the set of the 10 trials, each with 1000 particles. Also analyzed were the calculated values of the average percent captured of the 10 trials, and the standard deviation between these 10 trials. Figure 43 shows the recorded data from running 10 identical simulations with 1000 particles in each simulation.
Figure 43: Error Analysis of the 3-D Cylindrical Filter Simulations

Figure 43 shows that for each section of pipe there is similar standard deviation between the trials. Because each section’s capture efficiency in the cylindrical filter is dependent on every earlier section, if the standard deviation changes over the sections this implies that not enough samples were investigated and the “true” average (an average over an infinite number of particle) is not known. Since the standard deviation change is small and unchanging, this implies that using 10 tests of 1000 particles gives an accurate average with minimal statistical error. Also seen in Figure 43 is that the percent difference between the extreme values, minimal and maximum percent captured by a 1000 particle test, compared to
the average percent captured over the 10 tests was similar, which shows that the results are repeatable.

The percent captured in each section of the 10 tests were then combined to see the total percent captured by the filter. This was done to see how the statistical differences in each section combine to form a statistical difference in the overall capture efficiency of the filter. As was done with the individual sections, the overall filter capture efficiency maximum and minimum values of the 10 runs were recorded. The average and standard deviation of the overall capture efficiency of the runs were calculated.

Table 1 shows the maximum and minimum capture efficiencies of the filter with the average capture efficiency and the standard deviation between the 10 trials of 1000 0.1 μm diameter particles.

<p>| | |</p>
<table>
<thead>
<tr>
<th></th>
<th></th>
</tr>
</thead>
<tbody>
<tr>
<td>Average</td>
<td>45.87%</td>
</tr>
<tr>
<td>Minimum</td>
<td>42.9%</td>
</tr>
<tr>
<td>Maximum</td>
<td>47.9%</td>
</tr>
<tr>
<td>Standard Deviation</td>
<td>1.88%</td>
</tr>
</tbody>
</table>

Table 1: Error Analysis of Overall Filter Capture Efficiency with 10 Simulations of 1000 0.1 μm Particles

Table 1 shows that for each run done with 1000 particles of 0.1 μm diameter that the average calculated overall capture efficiency will vary from the “true” capture efficiency for the 0.1 μm diameter particle by 1.88%. This 1.88% difference between a typical simulation and “true” average corresponds to 4% of the “true” average. Therefore, any calculated overall...
capture efficiency based on 1000 particles is expected to be within 4% of its “true” value. Using a similar calculation and the data presented on Figure 43, the sectional capture efficiency based upon on 1000 particles is expected to be within approximately 9% of its “true” value.

3.2.3 Chemical Vapor Deposition

As mentioned in section 1.2, Brownian motion is of interest in the metal organic vapor phase epitaxy (MOVPE) of aluminum nitride (AlN). The MOVPE of AlN is performed in many different types of reactors, of which one type is a horizontal reactor. In these reactors, the reacting gases flow into the chamber and as they pass over the heated substrate, because of the chemical reactions, an epitaxial AlN film is grown on the substrate. However, in this process, micron and sub-micron sized particles of AlN have been observed to have formed as suspended solids in the gas (Creighton et. al., 2004). These particles are thought to form right over the top of the wafer (substrate) and have been seen to move away from the substrate quickly. It is widely believed in the literature that this is because of thermophoretic forces acting on the particle (Creighton et. al., 2007). This thermophoretic force is a force caused by the local temperature gradient. By carefully examining the analytical equation describing the movement of the Brownian particle, equation (4), the reason for the thermophoretic force can be seen. In this equation, it is seen that the particle movement is dependent on the temperature of the fluid at the particle. Since a particle is not a point mass as it is simulated, but rather a solid volume and takes up space, a fluid might have different properties at different location on the outside of the particle, and therefore, a different Brownian force on all sides of the particle. Therefore, if the carrier fluid has a temperature gradient at the location of the particle using a simple approximation of the Brownian motion by just using the fluid temperature at the particle center location is inaccurate. This approximation does not take into account the difference
between the temperatures on the different sides of the particles. Therefore, in order to properly model this situation, the thermophoretic force must be included.

The first chemical vapor deposition simulations were run in a horizontal reactor. This was done because the carrier fluid flow is the simplest, and therefore, the effects of the thermophoretic effects can be understood before more complex flows and more commercially utilized reactor geometries are simulated. Figure 44 shows a 2-D schematic geometry of the horizontal reactor used.

![Figure 44: 2-D Schematic of a 3-D Horizontal Chemical Reactor](image)

In this simulation, the mesh spacing was not equal, as more cells were used in the lower part of the y-direction (near the wafer and bottom of the reactor) than at the upper portion of the reactor. This was done because of the sharp temperature gradient in the y-direction. The setup and boundary conditions used is that of a common MOVPE reactor (Endres, 2010). A wafer temperature of 1000 K was assumed. An inlet flow rate of $2.6 \times 10^{-5}$ kg/s was used, which
corresponded to a maximum x-direction velocity of approximately 0.2 m/s. The fluid density was 1.1614 kg/m$^3$ and viscosity of 1.846x10$^{-5}$ kg/m-s. This gives a $Re_L$ of approximately 125, which is still in the laminar regime. For a 1 μm diameter particle, $Re_p$ is approximately 0.01.

As mentioned earlier, the first test done was to gain fundamental understanding of the effect of the thermophoretic force. This is accomplished by inserting particles at the inlet of the reactor and tracing the particle until it exits the reactor. This test shows that the thermophoretic force “pushes” the particle away from the heat source. This is seen to occur at 0.1m and end at 0.15 m; this is because this is where the heated wafer is. A time step of 1 ms was used. Figure 45 shows the trajectory of a 0.05μm particle with drag, Brownian forces and thermophoretic forces active.
It shows that there is clearly some Y-direction movement of the particle in the first 0.1 m of the reactor. This is due to the prescribed inlet boundary condition which has an inlet region. Then, beyond 10 cm, the thermophoretic causes the particle move away from the heated substrate. From this simulation it can be seen that the thermophoretic force appears to have a range from the wafer at which it affects the trajectory of the particle. This simulation is repeated with a numerous particles to determine the mean path. Figure 46 shows the average...
particle trajectory using 500 1 μm diameter particles with the same forces active and a time step of 0.01 s.

Figure 46: 1 μm Diameter Particle Average Trajectory in a Horizontal Reactor with Thermophoretic Forces Active for 500 Particles

This shows a very similar result as the 0.1 μm individual particle simulation. An interesting phenomenon noticed is that after the wafer, at 15 cm, the averaged particle location tends back towards the bottom of the reactor. This was noticed with the individual particle but
was attributed to Brownian forces. This cannot be the case in Figure 46 because 500 particle trajectories were averaged and Brownian motion should not be seen. Therefore something else must be causing this movement. The first effect investigated was the flow field at this location, but there was minimal y-direction velocity and this was not significant enough to cause the movement seen. The only other force active in this simulation is the thermophoretic force which requires a local temperature gradient in order to be active. Figure 47 shows the fluid temperature contours.

![Temperature Contour of a Horizontal Reactor](image)

Figure 47: Temperature Contour of a Horizontal Reactor

Figure 47 shows that because of the isothermal wall right after the heated wafer there is a temperature gradient seen in the fluid. However, the gradient is not near as strong as the gradient above the wafer. This temperature gradient is due to the flow heating up over the
substrate and then continuing downstream where the wall temperatures are kept isothermal at 300 K. Because this gradient exists, a particle that has moved past the heat source will either move towards the bottom or the top of the chamber due to thermophoretic forces from the temperature gradient.

The effects of the thermophoretic forces are investigated to see how this force could affect particles all over the simulation domain. Figure 48 highlights the effects of the thermophoretic forces by showing the average particle trajectory of 500 μm diameter particles at various starting locations of particles at the inlet.
This clearly shows that the particles closer to the wafer undergo more significant thermophoretic forces and for particles further away from the wafer the force essentially has no effect.

The same reactor geometry and boundary conditions are used, but now the particles that are inserted are the AlN particles that form as suspended solids above the wafer. This is simulated by solving the carrier fluid with chemical reactions turned on. In the process of simulating the growth of AlN complex gas-gas and gas-surface reactions are required to fully capture the process. The reactions used are the set of reaction as determined by Mihopoulos,
Gupta and Jensen. Figure 49 is a schematic of the reaction pathway for AlN grown (Mihopoulos et. al., 1998). Table 2 shows the specifics of the reactions considered in this model.

Figure 49: Schematic of the AlN Decomposition and Reaction Pathways (Mihopoulos et. al., 1998)
Using this model both AlN epitaxial growth and AlN particle formation is predicted. For the current simulations AlN particles with varying diameters are inserted above the wafer. The particle diameter was assumed to depend on the predicted concentration of AlN. For the current simulations the diameters vary from 0.1 µm to 2.3 µm, increasing based upon the predicted concentration. Figure 50 shows a contour plot of the predicted concentration levels of AlN particles. It is assumed from this plot that the AlN particles form above the substrate and are then carried downstream by the fluid. Once downstream, the particles diffuse over the entire height of the reactor.
Particles are inserted in various cell centers above the wafer and are given the fluid velocity at that location. These simulations use 400 particles in each run. Figure 51 shows the average particle trajectory as predicted by particles with varied diameter based upon starting location.
Figure 51: Varied Diameter AlN Particle Trajectories in a Horizontal Reactor

Figure 51 shows that the average particle is moved by the thermophoretic force very quickly away from the wafer. As noted by Creighton these AlN particles are unable to contribute to the AlN film growth because they are no longer near the wafer, thus wasting precursor gases, and potentially clogging downstream filters (Creighton et. al., 2007). The phenomenon shown in Figure 46, of the particle moving to the bottom because of the temperature gradient, is seen very clearly again by the AlN particle simulation in Figure 51. This predicts that the particles will
constantly bounce off of the bottom wall. In this simulated setup the particles continually bounce off the wall being carried downstream and out the outlet, and do not cause any problems. However, in a real experimental setup, this could cause an issue because these particles could form a coating on the wall.

A second situation where thermophoretic forces play a significant role is in the MOVPE of AlN in a vertical reactor. A vertical reactor has a more complex flow field due to a stagnation point in the fluid flow. These reactors are more commonly used as the growth rate of AlN tends to be higher than horizontal reactors. A simplified reactor model of the Thomas-Swan® vertical reactor, a commonly used reactor in commercial applications, is used. Figure 52 shows a 2-D schematic of the reactor.

Figure 52: Thomas Swan® 2-D Axis-Symmetric Reactor Schematic

Just as what was seen in the horizontal reactor, a temperature gradient in the fluid is observed, but because of the stagnation point in the center, the flow pattern is that of a three-
dimensional radial boundary layer that is suppressed. The temperature gradient near the wafer is very high. Figure 53 shows the temperature gradient relative to the reactor height, and magnified.

The geometry used was a 45° slice of the complete reactor, with symmetry conditions to on the two azimuthal planes. This means that the direction of travel for the particle to escape is in both the y and z directions (the radial direction). Figure 54 shows the trajectory of a single 0.05 μm particle inserted above the back half of the wafer with a zero initial velocity using 1 ms time steps.
Figure 54 shows the particle travels primarily due to the thermophoretic force and drag forces, as seen in the horizontal reactor and that Brownian forces effects are minimal compared to these forces. Simulations were also performed run to see how formed particles travel in the reactor, and for these simulations 400 particles were used at each starting location. Particles
were all inserted on the same 2-D plane, so that they started in the middle of the “slice,” so they would have the greatest distance to travel before interacting with the symmetry walls of the slice. Any interaction with the symmetry planes by a particle was treated as a particle interacting with a wall, and therefore an elastic and specular reflection was used. As with the horizontal reactor, particles were inserted based upon the simulated concentration of AlN particles and range from 0.1 µm to 2.5 µm, with increasing particle diameters as the insertion location is more downstream. These simulations were ran until the particles were beyond the substrate and then ended, using time steps of 1 ms. Figure 55 shows a zoomed-in version of the results of the simulations, where the location of the wafer starts at 0 m in the radial direction and the radial direction is a normalized distance based upon the wafer length.
Figure 55: Varied Diameter AlN Particle Average Trajectories in the Thomas Swan® Reactor for 400 Particles
Figure 55 shows a similar result to what was shown for formed particle trajectories in the horizontal reactor in Figure 51. The average particle quickly moves away from the wafer because of the thermophoretic force. Particles formed on the second half of the wafer are greatly influenced by the post substrate temperature gradient and are forced to the bottom of the chamber, continuously bouncing off the wall. Figure 56 shows an interesting result of the simulation and highlights this by showing an average particle introduced at the front edge of the wafer, with the carrier fluid flow velocities shown behind the particle trajectory.

Figure 56: 0.1 μm Diameter AlN Particle Average Trajectory in the Thomas Swan® Reactor for 400 Particles
The result seen is that the effect of the thermophoretic force is greater than the effect of the drag force caused by the fluid flow. This is apparent because the particle travels against the normal flow of the fluid. As the particle moves away from the heated substrate the temperature gradient becomes weaker, and therefore, the thermophoretic force is weaker, and the particle starts moving with the carrier fluid as expected.

For the MOVPE of III-V in chemical reactors, pulsing the incoming precursor gases has been proposed as a way to maintain growth while potentially limiting the growth of suspended particles (Hiramatsu et. al. 1996; Ofer and Galewski, 2001). The pulsing pattern consists of pulsing one precursor gas, then a hydrogen purge, then the other precursor gas, and then another hydrogen purge. Figure 57 depicts the proposed pulsing pattern.

![Figure 57: Mass Flow Rate Schematic for Pulsed CVD](image)
Because of the pulsing, the flow field can no longer be modeled as steady since the inlet conditions are changing periodically. This results in a complex flow pattern. The time averaged inlet mass flow rates of the gases over a complete pulse cycle are kept consistent with the steady mass flow rates. The pulsing results in smaller concentration of AlN particles in the gas phase. Nonetheless some particle formation still occurs right above the wafer. For this simulation, particles were inserted based once the concentration reaches a certain threshold value. The particle trajectories were then recorded.

The Brownian motion simulation was started near the beginning of the 10th cycle. The 10th cycle was chosen because after this time period, the flow reaches a quasi-steady state, and the time averaged AlN wafer deposition rate over a pulse cycle is steady. The location in the 10th cycle was chosen because particles are seen to form when either precursor gas pulse is occurring. It is assumed that the particle formation over the wafer is the same as it would be in any later pulsing cycle. A time step of 10 ms is used for the fluid solver, while a 1 ms step is used for the particle solver. The simulation is carried on for almost 3 complete pulse cycles. Figure 58 shows a trajectory of a single 1 µm started near the middle of the wafer.
Figure 58: 1 μm Diameter AlN Particle Trajectory in Unsteady Flow: Example 1

Figure 58 highlights two behaviors. First, the particle moves downstream and second, there is minimal movement away from the wafer. Because of this the Brownian motion becomes more noticeable than with the steady state simulations. The increase in observed Brownian motion is due to two reasons, the first being that there is a smaller downstream velocity for most of the pulse cycle and second the thermophoretic is weak. The smaller
downstream velocity causes the particle to take longer to move downstream and is explained by the changing flow rate into the reactor. The reduced thermophoretic force is most likely attributed to the thermal field not being fully developing because of the amount of time into the simulation and flow carrying the heated fluid downstream. Figure 59 below shows another particle trajectory that, over the tracked time, moves a relatively smaller distance away from the wafer.

Figure 59: 1 μm Diameter AlN Particle Trajectory in Unsteady Flow: Example 2
Figure 59 shows that this particle travels a smaller distance downstream over the same time duration when compared to the particle in Figure 58. This highlights an effect and that is the AlN particles for pulsed CVD stay over the area of the wafer longer. Because of this and Brownian motion the particle is able to move towards the wafer and, as shown with the particles tracked, comes very close to the wafer. From an applications standpoint, this may be beneficial, since it may allow incorporation of the AlN particle into the film.

Comparing Figure 58 and Figure 59 also shows that the particles take very different trajectories based on the distance away from the wafer. The different trajectories can be predicted by earlier examples shown, but was not observed because for the exact same starting conditions a large variance in the distance away from the wafer is not seen. Further, this effect is not observed during steady state CVD and is therefore due to the unsteady flow fields and the time dependent conduction of heat from the wafer. The weak thermophoretic forces seen could be because of the initial conditions of the chamber and the heat slowly moving from the heated substrate to the gas. Even though particle AlN formation and film growth has reached a quasi-steady state, the thermal field may not have yet. Therefore, under this operating condition, the Brownian force becomes more important compared to the steady state conditions.

This simulation was repeated with only a few modifications. The starting location moved to the front of the wafer and a 0.1 µm diameter particle was tracked. The simulation is continued for only 1.5 pulse cycles after the particle is inserted. Figure 60 shows the resulting trajectory of a 0.1 µm diameter particle inserted on the front edge of the wafer.
Figure 60 shows a different result than seen for the steady state case, Figure 56, with the particle not tending away from the wafer at first. This further confirms that while there will be AlN growth, there might not be a fully developed thermal field. However, this also shows that the Brownian motion of the particle becomes very important and might result in the AlN
particle adhering to the wafer surface, possibly enhancing the growth rates. Figure 61 shows a trajectory of a particle that gets close to bouncing off of the wafer, but then is sharply forced away multiple times. A particle like this might break down when it gets near the high temperature region near the wafer and deposit, adding to the film growth.

Figure 61: 0.1 μm Diameter AlN Particle Formed above Wafer Front Edge Trajectories in Unsteady Flow: Example 2
4 Summary and Conclusion

The random forces that cause Brownian motion are clearly very powerful forces that affect the trajectory of micron-size particles. In this study a general-purpose Brownian motion numerical model was developed. This model is based on the Langevin equation. The resulting ordinary differential equations are solved by using the Newton-Raphson method after backward Euler discretization. In order to track the particle’s trajectory through the background fluid, a three-dimensional (3-D) tracking algorithm was developed. The tracking algorithm and particle motion solver are applicable to arbitrary two-dimensional (2-D) and 3-D geometries discretized using unstructured mesh topology. The model was fully coupled to the carrier fluid flow solver, allowing for unsteady flows to be simulated. To apply this model to various applications of interest, viscous drag, Brownian forces, lift, and thermophoretic forces were considered. The Brownian motion model was first validated against analytical results and then explored for different applications. The above mentioned phenomena were investigated on three different applications; H-filters, cylindrical filters, and chemical vapor deposition (CVD) of Aluminum Nitride (AlN) film in a CVD reactor.

In this work, both the H-filter and cylindrical filters were designed to optimally filter out desired particles by studying the affect of modifying the carrier fluid’s viscosity, temperature and the desired particle diameter. It was shown that for the same carrier fluid a particle with a
smaller diameter is more likely to be filtered. The fluid properties were then varied as the
diameter was held constant, and it was shown that with either a higher fluid temperature or a
lower fluid viscosity that the filter efficiency increased. The filter applications demonstrated
that Brownian motion can be utilized to filter out particles.

Also studied in this work was the effect of Brownian motion on AlN particles formed
during the CVD of AlN film. Steady CVD was first investigated. It was found that the drag and
thermophoretic forces primarily dictate the particle trajectories in both horizontal and vertical
reactors. For the vertical reactor, the CVD simulations were repeated using pulsed incoming
gases (pulsed CVD). The particles were inserted above the wafer with a particle diameter based
on the AlN particle concentration level after the system reached quasi-steady state. The particle
trajectories in the unsteady flow showed significantly different behavior than steady CVD.
These particles exhibited less effect of thermophoretic forces, and therefore, the Brownian
forces on the particle appeared dominant. It was seen that the particles in the unsteady CVD
case may come in contact with the epitaxial film (wafer surface). Therefore, the Brownian
motion of formed AlN particles in a pulsed CVD application is important when predicting particle
trajectory.
Further work is necessary to expand the scope of the current model and make it more universally applicable. In order to accomplish this, particle-particle interaction should be considered. To do this, the model would need to be modified so that several particles are tracked simultaneously and particle-particle short-range interaction forces would need to be added to the equation of motion of the particle. Also particle-particle collisions would need to be modeled. Another interesting enhancement would be to enable two-way coupling between the carrier fluid flow solver and the Brownian motion solver. This would allow the model to accurately simulate applications where the particle diameter is comparable to the characteristic length of the device being modeled.

Finally there is a strong need for performing carefully designed experiments, which would generate data that could be used for validation of the simulated results.

5 Future Work
6 References


