Parallel solution of Uni-directional coupled PBM-DEM for fast evaluation

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Abstract

abstract text goes here ...

Keywords: Population balance model, heteroaggregation, alginate, chitosan, oppositely charged

1. Introduction & Objectives

partiulate proceses and importnatce

Half of all industrial chemical production is carried out using particulate processes (Seville et al. (1997)). Common particulate processes in industry include crystallization, granulatioin, milling, polymerization (cite https://www.hindawi.com/journals/mse/2013/475478/). Important products that are produced using particulate processes include detergents, aerosols, fertilizers, and pharmaceuticals. A particularly important process to the pharmetucitcal industry is granulation.

could mention granulation and its microscale behavior by which mean mechanisims so that raeder knows what we are refering too and knows we have a focus on granulation etc kind of kills two birds with one stone. Then segway to complexity of these systems is preseverved which preserves segway into other modeling stuff. also good mention induistry aim to tailor products to meet very exact needs then will set up importantce of next paragraph

difficutl physics, cost to industry, and need for models

Granulation is a particle design process used to control the size, chemical composition, and other pertiant particle characteristics to the end product. Granulation is comprised of several rate processes - nucleation, aggregation, and breakage. These rate processes are dictated by the microphenomena that result from the particle-particle interactions of the system which are chaotic making efforts to model them very difficult. The diffculties of modeling these processes is compunde in the pharma industy due to their high quality control standads they must adhere to. Barrasso and Ramachandran (2015) these microscale phenomena are what make particulate processes extremely complext/difficult to develop physical models for. In general particlate processes have consider merging next paragraph here or so.

Particulate processes have complex phenomena due to the chaotic microscale behaviour of individual particles in the system that determine the bulk behaviour of the system. Due to the complex nature of these processes there is a lack of governing equations which can accurately describe them (Sen et al. (2013)). As a result industries such as pharmaceuticals use expensive heuristic studies and inefficient operational protocols with high recycle ratios to meet strict regulatory standards (Ramachandran et al. (2009)). In an effort to cut costs and improve efficiencies there is a high demand for accurate models that can be used for process design and control systems.

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intro DEM and PBM with breif concept explaination - hammer on how long either model takes to run and how that an issue

Discrete element methods (DEM) have proven to be an accurate way of model a bulk material properties of particulate systems using microscale particle level interactions way modeling microscale part-part behavior that develops into bulk properites of particulate system. DEMs use newton's equations of motion to model each particle in they system of study and its interactions with the system geometry and othe particles which is what enables DEMs to model the small scale phenomena that determines the bulk behavior of particulate systems. The downside to DEMs is that they take large amounts of time to solve since they model each particle of the system. Often times scientists need to alter simulation properties to make DEMs run faster but it makes them less accruate (CITE NUMEROUS DEM WORKS WE LATER USE and in chai paragraph from intro). Another more computationally efficient, but less accurate technique, used for these systems are population balance models (PBM). PBMs are semi-mechanistic, meaning they use population averages and probablites to capture more bulk behavior with little computation but still seak to try to capture some of the microphenomena of particle-particle interactions, but in less detail than DEM. Even though PBMs are much faster than DEMs they still can take a significant amount of time to solve (CITE NUMEROUS WORKS ON PBMs WE USE LATER and from chai work in intro).

coupled PBM-DEM model same stuff as for pbm or dem above - still hammer on that they take too long

In attempts to take advantage of the highly accurate modeling capabilites of DEMs and the efficiency of PBMs there has been great interest in hybrid PBM-DEM methods. The typical work flow of these PBM-DEM coupled models involves using short DEMs to capture the particle-particle level interactions of the system and then those particle-particle physics from the DEM are fed into the PBMs particle-particle so that the PBM can more accurately simulate bulk system behavior (CITE NUMEROUS coupled DEM-PBM people we cite later on and from chai paragraph in intro). Despite the performance benefits of these coupled PBM-DEM models they still take too long to solve for optimization, controls, design, hundreds of sims but each sim take hrs to evaluate ddd not practical.

parallel computing intro, need, how if fit into this problem - answer to those things taking too long!

In the past when faced with computational heavy tasks scientists have used parallel computing to distribute large workloads across many computers which work together to solve the problem more quickly. Parallel computing as been used to speed up list stuff here cfd, MD, finite diffs, etc, even PBM!. To solve a problem in parallel it needs to be possible to break the problem down into smaller pieces which can be solved simultaenously across many compute cores and then those solutions need to be able to be reassembled for a total solution perhaps explain Pllcomp in general with one setnance is not can of worms I want to open - reader could go figure it out themselves if they like. To solve problems in parallel computing clusters are often used and have been dropping signifcantly in price making them more accessible than ever want to show that cheaper and user can get excite but might need to throw some ideas about cloud access or amazon bs or something?. A cluster is essentially a collection of conventional computers connected together via a high speed network like ethernet or infiniband. In the past textcolorcyanexamples of speed up gains from using parallel computing so solve stuff so reader gets ideas about what coudl do. this makes good segway into talking about the potentail opportunites afforded by using these methods to solve these problems.

potential benefits this could have 1. unprecedented accuracy 2.wide adoption of using these models since they are so fast 3. cheaper drugs 4. MPC accurate but in real time 5. parameter

estimation for QbD 6. etc.

1.1. Objectives

The main objective of this study was to develop new techniques to parallelize our model and use high perfomance computing (HPC) equipment in order to reduce the computation times for the linked DEM-PBM simulations. Specific objectives of each individual study are listed below:

- Developed a 4-Dimensional PBM that is parallelized using hybrid techniques (MPI + OpenMP) for optimal utilization of modern high performance computing equipment.
- LIGGGHTS® has been used to perfrom Discrete Element Model (DEM) simulations to model the micromechanics of the Lodgitech granulator.
- These 2 techniques were linked uni-directionally to provide an accurate model of the granulator.

2. Background & Motivation

2.1. Particulate Processes

Particulate processes more details that may be useful that didnt get into in intro

Particulate processes are ones in which a system of discrete species exist, such as granules or catalyst pellets, that undergo changes in average composition, size, or other pertinent properties. Such processes are prevalent in the pharmaceutical industry. These processes have great oppurtunities in better equipment design, process efficiency and scale-up (Ketterhagen et al. (2009)).

specific part proc for use granulation and the mechanisims of granulation?

cost to industry and issues with industry methods curretnly

QbD / models the answer to their problems!

talk about how its so hard/ impossible to model them thoguht - make them worried about the problem at hand

then we talk about how we are going to solve that modeling problem in this curretn paper!

The quality by design (QbD) concept i.e, theoritically modelling the process for better quality of the product is being implemented by the pharmaceutical industry due to its cost benefits. Also, the paradigm shift of the industry towards continuous manufacturing, emphasizes the need to model these processes more accurately. This further helps develop better control stratergies for the process.The modelling of these processes is more time consuming and computationally expensive when compared to a fluid systems since the particles are considered as individual entities rather than a continuum like in fluid systems. The models discussed represent the particle-particle interaction at meso and micro-scale.

after talkng about how impactful QbD would be talk about physics that makes these part systems so hard to model/ develop govning eqns for. this will be good segway into the modeling section since now they know what needs to be considered kind of and what difficulties are in modeling etc.

2.2. Modeling

2.2.1. Discrete Element Modeling (DEM)

Discrete Element Method is a simulation technique used to monitor the behaviour of each particle as a separate entity compared to other bulk continuum models. This method tracks the movement of each of the particles with in the space, records the collisions of each particle with

the geometry as well as with each other and it is also subject to other force fields like gravity (Barrasso and Ramachandran (2015)) This model is based on the Newton's laws of the motion and is expressed as in Equations 1 and 2 :

$$m_i \frac{dv_i}{dt} = F_{i,net} \tag{1}$$

$$F_{i,net} = F_{i,coll} + F_{i,ext} \tag{2}$$

In the above equations m_i represents the mass of the particle, v_i represents the velocity of the particle, $F_{i,net}$ represents the net force on the particle, forces on the particle due to collisions and other external forces are represented in $F_{i,coll}$ $F_{i,ext}$ respectively.

The distance between each particle calculated at every time step and if the distance between two particles is less than the sum of the radii (for spherical particles) a collision between the two particles is recorded. The tolerance for overlap is low in the normal as well as the tangential direction (Cundall and Strack (1979)). Microscale DEM simulations are computationally demanding and simulations may take upto several days to replicate a few seconds of real time experiments. Many methods have been implemented to increase the speed of these simulations, such as scaling by increasing the size of the particles. These approximations are good in understanding the physics of the system but are not directly applicable to process-level simulations.

Thus, this method for granular powder is usually replaced by Population Balance Method which is a much quicker approximation as it is a bulk model.

2.2.2. PBM

Population balance models (PBM) predict how groups of discrete entities will behave on a bulk scale due to certain effects acting on the population with respect to time (Ramkrishna and Singh (2014)). In the context of process engineering and granulation, population balance models are used to describe how the number densities, of different types of particles, in the granulator change as rate processes such as aggregation and breakage reshape particles (Barrasso et al. (2013)). A general form of population balance model is shown here as equation 3.

$$\begin{aligned}\frac{\partial}{\partial t}F(\mathbf{v},\mathbf{x},t)+\frac{\partial}{\partial \mathbf{v}}[F(\mathbf{v},\mathbf{x},t)\frac{d\mathbf{v}}{dt}(\mathbf{v},\mathbf{x},t)] + \frac{\partial}{\partial \mathbf{x}}[F(\mathbf{v},\mathbf{x},t)\frac{d\mathbf{x}}{dt}(\mathbf{v},\mathbf{x},t)] \\ = \Re_{formation}(\mathbf{v},\mathbf{x},t) + \Re_{depletion}(\mathbf{v},\mathbf{x},t) + \dot{F}_{in}(\mathbf{v},\mathbf{x},t) - \dot{F}_{out}(\mathbf{v},\mathbf{x},t)\end{aligned} \tag{3}$$

In equation (3), $\mathbf{v}$ is a vector of internal coordinates. For modelling a granulation process $\mathbf{v}$ is commonly used to describe the solid, liquid, and gas content of each type of particle. The vector $\mathbf{x}$ represents external coordinates, usually spatial variance. For a granulation process this account for spatial variance in the particles as they flow along the granulator.

PBM extensively used for calculating granulation rate processes (e.g. aggregation, consolidation and breakage) but it cannot provide information related to mechanistic kernels of these rate processes i.e. collision frequency, collision efficiency, particle velocity etc. (Sen et al. (2014)). On the other hand, DEM able to determine these entities. A hybrid model for one-way coupling has been reported for continuous mixing (Sen et al. (2013) & Sen and Ramachandran (2013)) and is discussed in later section.

talk about kernels - empirical, semi mech, and DEM informed? talking about kernels then mentioning there is DEM informed could be GREAT SEGWAY to multi-physics models dotdotdot combined PBM DEM etc.

2.2.3. Multi-physics models

The goal of mutli-physics or coupled models is to try get the best of both worlds. Someones want faster modeling of one or the better accuracy of others. It is somewhat dependend on the goal of the work and nature of the over all context of the problem being solved (industry wants faster over more fundemental etc).

Due to the potential benefits a coupled PBM-DEM model could have there has been a number of studies of the topic. talk about past works here. pros cons good bad potential benefits tricks methods. mention stuff about difficulties in getting the the two models to talk to each other etc.

The use of multi-physics models has been used to understand the behaviour of particle systems. These models help understand the physics of the system at various scales *iė* micro, meso and macro scale(Sen et al. (2014)). Particle process dynamics have been inferred from coupling of various physics models *viz.* Computational Fluid Dynamics (CFD), DEM and PBM. Earlier works from Sen et al. (2014) and Barrasso and Ramachandran (2015) have successfully predicted process dynamics of the granulation process.

closing remarks about this paper in regards to these other coupled works? IN LAST SENTENCE OF THIS SECTION TOUCH ON HOW LONG IT CNA STILL TAKE EVEN WITH THIS BETTER METHOD OF DOING IT this will be great segway back into parallel computing with detail this time!

In this work, a coupling of DEM and PBM was implemented. The PBM gives meso scale information and the DEM gives particle scale information. Combination of these two methods help comprehend the process dynamics with more accuracy. The calculations involved due to the number of particles involved in the DEM process as well as PBM become very computationally heavy. The recent development in the design of CPUs and increasing number of cores in the CPU, it makes sense to utilize them to make the simulations faster. Thus, implementation of various parallel computing techniques was employed in this work to help improve the simulation times.

2.3. Parallel Computing and Computer Architectures

The goal of parallel computing is to distribute large amounts of computation across many compute cores to solve problems faster (Wilkinson and Allen (2005)).

2.3.1. Computer Architecture

Parallel programs are commonly run on computer clusters. Computer clusters are a collection of nodes interconnected by a high speed communication network for message passing from one node to another. Analogously to a conventional PC each node as one or more CPUs and RAM. Commonly nodes are manufactured with two CPUs, each CPU is a multi-core meaning it has multiple compute cores that each can carry out calculations separately from one another. CPUs also have built in memory called cache that is much faster than RAM which is why for optimal performance cache utilization should be favoured over RAM when possible. On a node memory is divided by CPU sockets, so each CPU has direct access to memory local to its own socket, but accessing memory on another socket is much slower Jin et al. (2011). For this reason data that is needed for computation should be stored locally to the CPU that needs it.

chai did decent rewording here but did change some of the meaning so will leave here for reference later on as I go through versions. Two common classes of computer architecture classified by memory locality features are distributed memory systems and shared memory systems. These two classes co-exist in a cluster, thus providing the benfits of each. All the nodes share memory using explicit message passing while each has its own independent memory. The cores on each node can access data from the shared memory without any explicit message passing statements

from the user. While designing a parallel program all these aspects need to be considered for optimal perfomance of the code (Adhianto and Chapman (2007)).

Computer architectures are often classified by memory locality features. There are two distinct classes, distributed memory systems and shared memory systems. A cluster is a combination of the two classes. Each node operates its memory independently of the other nodes and explicit message passing is needed to share memory between nodes. While the cores on a node can operate in shared memory mode since memory updates can be made without explicit message passing statements from the user. All of these aspects of the computer architecture should be considered when designing a parallel program for the best performance should reword better to make more like CACE paper. also not really the difference in shared and distributed memory especailly not from comp eng point of view.

2.3.2. Parallel Application Program Interfaces

Message Passing Interface (MPI) is a common parallel computing application interface standard. MPI is used for distributed memory parallel computing, this is because MPI will operate every MPI process as a discrete unit that does not share memory with the other processes unless explicit message passing is used. Even on shared a single node where the hardware is supports shared memory computing, MPI will still operate it in a distributed memory fashion Jin et al. (2011). Operating all cores as distinct units also means they each need their own copy of all variables used for computation which results in a large overall memory foot print compared to a same system if it was operated in shared memory.

Open Multi-Processing (OMP) is another application program interface stand for parallel computing. OMP is used for shared memory and can take advantage of shared memory systems which can result in much faster computation. It does not work well on distributed systems though. This prevents it from being used to efficiently carry out computations across multiple nodes of a cluster simultaneously Jin et al. (2011).

Since MPI is best for distributed computing and OMP is better for shared computing many individuals have studied the performance of MPI vs MPI+OMP methods and many studies have used MPI+OMP for scientific computation for improved performance. Often times a trade of is made between optimizing a program for performance and trying to make it flexible enough to run on many different computer architectures might need reference for this. A summary of some works addressing MPI+OMP methods for scientific computing and architecture features and concerns can be found in Bettencourt et al. (2017). In the conclusion to the work by Bettencourt et al. (2017) it was found that hybrid methods for PBMs allow the code flexibility for different architectures while still maintaining good performance. should I mention load balancing techniques of gunawan paper?. In the work of Bettencourt et al. (2017) only the external(spatial) coordinates of the PBM were parallelized. In this current work external and internal(compositions) calculations are parallelized. comment about how int and ext pll methods means better model for xyz reasons dotdotdot helps to explicitly say reason

3. Methods

3.1. DEM

3.1.1. LIGGGHTS

LAMMPS Improved for general granular and granular heat transfer simulations (LIGGGHTS® v3.60) (Kloss et al. (2012)) developed by DCS computing was used for all the simulation performed in this study. Edits were made to the compute_contact_atom source file to obtain particle − particle collisions. The aforementioned version of LIGGGHTS was compiled using the mvapich (mvapich2

v2.1) and intel (intel v15.0.2) compilers with the -o3 optimisation option as well as an option to use OpenMP threads was implemented. The speed improvements and the disadvantages are illustrated in Table (refer to the speed table). The studies were performed on STAMPEDE supercomputer located at Univeristy of Texas, Austin. The hardware configuration of each node consists of 2 8-core Intel Xeon E5-2680 processors based on the Sandy Bridge architecture, 32 gb of memory with QPI interconnects at 8.0 GT/s PCI-e lanes.

3.1.2. Geometry

In this study, the Lödige CoriMix CM5 continuous high shear granulator has been studied. Its geometry was developed using the SolidWorks^{TM} (Dassault Systèmes). This granulator consisted of a high speed rotating element enclosed within a horizontal cylindrical casing. The casing (shown in Figure 1) consists of a cylinder with diameter of 120 mm at the inlet and 130 mm at the outlet and having a total length of 440 mm. A vertical inlet port is provided at one end of the casing and an angled outlet port is provided at the larger end of the case.

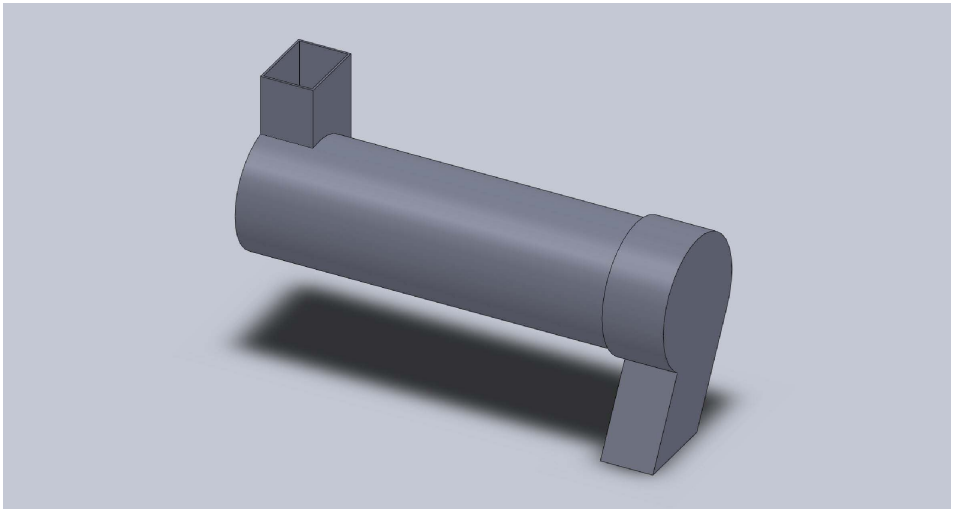


Figure 1: Shows an isometric view of the granulator casing.

The impeller consists of a cylindrical shaft of length 370 mm and diameter 68 mm with four flattened sides 15 mm wide running along the axis. The blades are placed on these flattened sides as shown in figure 2. There are three different blade elements on the shaft (figure 2). At the granulator inlet, there are 4 paddle shaped feed elements following which there are 20 tear drop shaped shearing elements and finally 4 trapezoidal blades near the exit. All these elements are placed in a spiral configuration. The final configuration of the granulator is shown in figure 3.

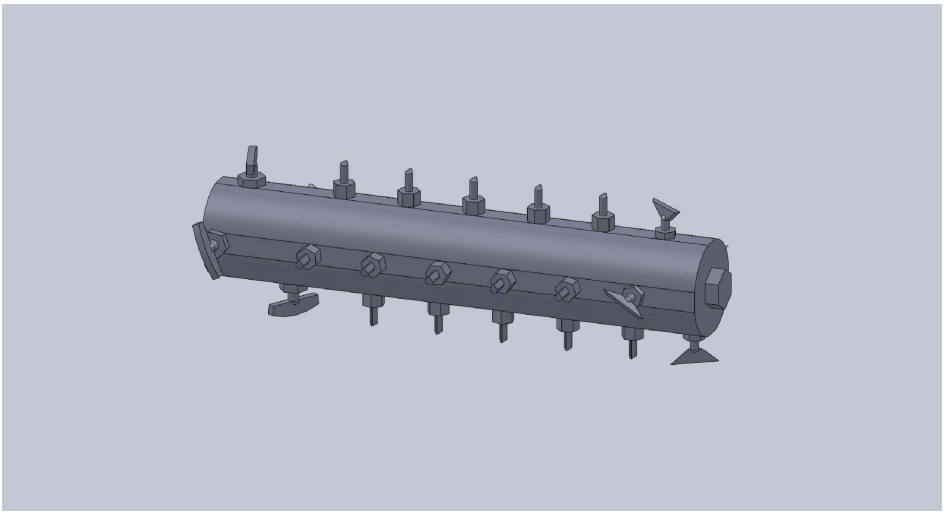
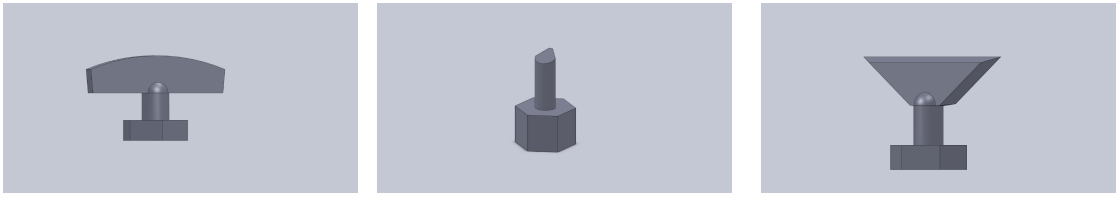


Figure 2: a) Shows the side view of the impeller. b) and c) show the front and back views of the impeller which correspond to the inlet and outlet ends respectively.



(a) Feed element **(b)** Shear element **(c)** Exit element

Figure 3: Various components of the impeller

3.1.3. Meshing

After the geometry was built in SolidWorks^{TM} (Dassault Systèmes) the shell and impeller were exported as STL files. The coarsest output option was used to keep the STL files small and simple for faster computations times. They were also exported not keeping there original coordinates This resulted in the impeller having 2802 faces and 1281 points with approximately a file size of 775 KBs. The shell had 1948 faces and 720 points and size was about 544 KBs.

Meshlab was used to align the STL files for importing into LIGGGHTS®. No mesh treatments were used on the STLs.

The meshes were then imported into LIGGGHTS® using the write command. This resulted in 50 elements of the impeller file having "highly skewed elements", which have more than 5 neighbours per surface or have an angle less than 0.0181185 degrees, that according to LIGGGHTS® would degrade parallel performance. The write exclusion list command in LIGGGHTS® was used and this exclusion list file as then used in the simulation to skip the highly skewed elements during the simulation

look at notes from olgitdraft pdf file on this section overall looks good though

3.1.4. DEM input file settings

The DEM simulation in LIGGGHTS® are setup using an input script which defines the physical parameters of the particles, importing of the geometry, particle insertion commands, various physics models to be used during the simulation as well as various compute and dump commands to help

print the data required for post-processing of the data. A script was prepared each of the particle was considered granular in nature. The Hertzian model was used for non-adhesive elastic contact between the granular particles. The particles were inserted inside the granluator from the inlet at a constant mass flow rate of 15 kilgrams per hour. The rotation speed of the impeller was kept throughout the study at 2000 rotations per minute. Such a high rotation speed was chosens since this would lead to high shear between the particles and the walls of the shell resulting in better size control of the granules. There were 2 sets of simulations that were performed, one with mono-sized partcles and second consisting of a distribution of sizes. The particle radii chosen for mono-sized simulation varied 0.59mm - 2mm, consecutive particles radii had volume twice of one before them. The radii range of the distributed size simulation was 1mm - 3mm. The difference in the mechanics of these two simulations is dicussed later in the results section. The physical constants used for the simulations are given in Table 1.

The simulation data was collected after a constant number of iterations for the visulation of the particles inside the shell, further post processing and to be used in the PBM. The collisions between each of the particles and the collisions between of the particle and the geometry was also collected. might be better to include more tables or more of the DEM settings info in the table like runtime time steps etc just as a nicer summary of thise sections work.

Table 1: Physical Properties of the particle for the LIGGGHTS® input script

Parameter	**Value**	**Units**
Young's Modulus of particles	8×10^6	$N.m^{-2}$
Young's Modulus of Geometry	1×10^9	$N.m^{-2}$
Poisson's Ratio	0.2	–
Coefficient of restitution (constant for all collisions)	0.4	–
Coefficient of static friction	0.5	–
Coefficient of rolling friction	0.2	–
Density of the granules	500	$kg.m^{-3}$

3.1.5. DEM data post processing

The post processing of the data obtained from the DEM simulations was done using MATLAB®. The first test run on the output data was to determine if the simulation had reached steady-state. The mass inside the granulator was found out by averaging it over 5 time steps and then compared to mass inside the granulator after every 10000 time steps (about 5x10^{-4} seconds) with a tolerance of about 10%. If the mass was found to be constant for most of the iterations, it was considered to be at steady state. Another test to determine steady state was to monitor the number of collision inside the granulator. It can be seen that the number of collision start to oscillate around a mean value. The number of collisions were then plotted and steady state time was determined. 1. could this be an issue if error is 10 percent that is large. what if we start form non steady state time bec need to consider if 10 of total mass is coming in in how much time for that to be fair right? if less than 10 percent comes in in that time or more then it will skew anaylsis? 2. can we make a multi plot thing that has time on x and other parameter in y. then show how collisions, mass flow, etc align at some time approximately which kind of indicates steady state or a constant flow has been reached. (since not supposed to use steady state in particulate systems apperently)

A precautionary script was also run so as to determine that no particles were lost due to overlap of the geometry with the particles as well as from particle particle overlap.

3.2. PBM

3.2.1. Model Development

The main PBM equation developed for this work can be expressed as shown below:

$$\frac{d}{dt}F(s_1,s_2,x) = \Re_{agg}(s_1,s_2,x) + R_{break}(s_1,s_2,x) + \dot{F}_{in}(s_1,s_2,x) - \dot{F}_{out}(s_1,s_2,x) \tag{4}$$

citation?

where, $F(s_1,s_2,x)$ is the number of particles with an API volume of s_1 and an excipient volume of s_2 in the spatial compartment x. The rate of change of number of particles with time in different size classes depend on the rate of aggregation $\Re_{agg}(s_1,s_2,x)$ and the rate of breakage $\Re_{break}(s_1,s_2,x)$. Also, the rate of particles coming into, $F_{in}(s_1,s_2,x)$ and going out, $F_{out}(s_1,s_2,x)$ of the spatial compartment due to particle transfer affect the number of particles in different size classes. The rate of change of liquid volume is calculated using the equation:

$$\begin{aligned}\frac{d}{dt}F(s_1,s_2,x)l(s_1,s_2,x) =& \Re_{liq,agg}(s_1,s_2,x) + \Re_{liq,break}(s_1,s_2,x) + \dot{F}_{in}(s_1,s_2,x)l_{in}(s_1,s_2,x) \\ &- \dot{F}_{out}(s_1,s_2,x)l_{out}(s_1,s_2,x) + F(s_1,s_2,x)\dot{l}_{add}(s_1,s_2,x)\end{aligned} \tag{5}$$

where, $l(s_1,s_2,x)$ is the amount of liquid volume in each particle with API volume of s_1 and excipient volume of s_2 in the spatial compartment x. $\Re_{liq,agg}(s_1,s_2,x)$ and $\Re_{liq,break}(s_1,s_2,x)$ are respectively the rates of liquid transferred between size classed due to aggregation and breakage. $l_{in}(s_1,s_2,x)$ and $l_{out}(s_1,s_2,x)$ are respectively the liquid volumes of the particles coming in and going out of the spatial compartment. $l_{add}(s_1,s_2,x)$ is the volume of liquid acquired by each particle in the compartment at every time step due to external liquid addition.

Similarly, the rate of change of gas volume is calculated using the following equation:

$$\begin{aligned}\frac{d}{dt}F(s_1,s_2,x)g(s_1,s_2,x) =& \Re_{gas,agg}(s_1,s_2,x) + \Re_{gas,break}(s_1,s_2,x) + \dot{F}_{in}(s_1,s_2,x)g_{in}(s_1,s_2,x) \\ &- \dot{F}_{out}(s_1,s_2,x)g_{out}(s_1,s_2,x) + F(s_1,s_2,x)\dot{g}_{cons}(s_1,s_2,x)\end{aligned} \tag{6}$$

citation?

where, $g(s_1,s_2,x)$ is the gas volume of each particle with API volume of s_1 and excipient volume of s_2 in the spatial compartment x. $\Re_{gas,agg}(s_1,s_2,x)$ and $\Re_{gas,break}(s_1,s_2,x)$ are respectively the rates of gas transferred between size classed due to aggregation and breakage. $g_{in}(s_1,s_2,x)$ and $g_{out}(s_1,s_2,x)$ are respectively the gas volume of the particles entering and leaving the spatial compartment. $g_{cons}(s_1,s_2,x)$ is the volume of gas coming out of each particle in the compartment at every time-step due to consolidation of the particles.

The rate of aggregation, $\Re_{agg}(s_1,s_2,x)$ in Equation 4 is calculated as

$$\begin{aligned}\Re_{agg}(s_1,s_2,x) =& \frac{1}{2}\int_0^{s_1}\int_0^{s_2}\beta(s_1',s_2',s_1-s_1',s_2-s_2',x)F(s_1',s_2',x)F(s_1-s_1',s_2-s_2',x)ds_1'ds_2' \\ &- F(s_1,s_2,x)\int_0^{s_{max_1}-s_1}\int_0^{s_{max_2}-s_2}\beta(s_1,s_2,s_1',s_2',x)F(s_1',s_2',x)ds_1'ds_2'\end{aligned} \tag{7}$$

citation?

where, the aggregation kernel, $\beta(s_1,s_2,s_1',s_2',x)$ is expressed as a function of collision frequency (C) and collision efficiency (ψ)

$$\beta(s_1,s_2,s_1',s_2',x) = \beta_o C(s_1,s_2,s_1',s_2')\psi(s_1,s_2,s_1',s_2',x) \tag{8}$$

where, β_o is aggregation rate constant.

Collision frequency is a function of particle size and is calculated from the number of collisions between group of particles obtained from LIGGGHTS®. A recent study shows that collision frequency depends on PSD as well (Sen et al. (2014)). Collision frerquency can be expressed as:

$$C(s_1, s_2, s_1', s_2') = \frac{N_{coll}(s_1, s_2, s_1', s_2')}{F(s_1, s_2)F(s_1', s_2')\Delta t} \tag{9}$$

In equation(9), N_{coll} is the number of collision between two solid particles in time interval Δt. The ψ in equation (8) can be expressed asvi

$$\psi((s_1, s_2, s_1', s_2') = \begin{cases} \psi_0, & LC((s_1, s_2) \geq LC_{min} \ \ or \ \ LC((s_1', s_2') \geq LC_{min} \\ 0, & LC((s_1, s_2) < LC_{min} \ \ or \ \ LC((s_1', s_2') < LC_{min} \end{cases} \tag{10}$$

In above equation, LC is the liquid content of particles and LC_{min} stands for minimum liquid content required for coalescence of particle.

Similarly, the breakage rate is expressed as-

$$\Re_{break}(s_1, s_2, x) = \int_0^{s_{max_1}} \int_0^{s_{max_2}} K_{break}(s_1', s_2', x)F(s_1', s_2', x)ds_1'ds_2' - K_{break}(s_1, s_2, x)F(s_1, s_2, x) \tag{11}$$

citation?

where, the breakage kernel $K_{break}(s_1, s_2, x)$ is formulated as

$$K_{break}(s_1, s_2, x) = C_{impact} \int_{U_{break}}^{\infty} p(U)dU \tag{12}$$

citation?

The rate of increase of liquid volume of one particle, $\dot{l}_{add}(s_1, s_2, x)$ is expressed as $\frac{(s_1+s_2)(\dot{m}_{spray}(1-c_{binder})-\dot{m}_evap)}{m_{solid}(x)}$ where, $(s_1 + s_2)$ is the total solid volume of the particle; $\dot{m}_spray$ is the rate of external liquid addition, c_{binder} is the concentration of binder in the external liquid (which is assumed to be zero in this case as pure liquid is added); $\dot{m}_{evap}$ is the rate of evaporation of liquid from the system (which is also assumed to be zero in this case) and m_{solid} is the total amount of solid present in the compartment.

The rate of decrease in gas volume per particle due to consolidation is calculated using the following expression:

$$\dot{g}_{cons}(s_1, s_2, x) = c * (\nu_{impeller})^{\omega} * V(s_1, s_2, x)\frac{(1-\epsilon_{min})}{s}[g(s_1, s_2, x) + l(s_1, s_2, x) - (s_1 + s_2)\frac{\epsilon_{min}}{1-\epsilon_{min}}] \tag{13}$$

where, c and ω are the consolidation constants; $v_{impeller}$ is the impeller rotational speed; $V(s_1, s_2, x)$ is the volume of particle, ϵ_{min} is the minimum porosity; $g(s_1, s_2, x)$ and $l(s_1, s_2, x)$ are respectively the gas and liquid volume of the particle.

Particle transfer rate, $F_{out}(s_1, s_2, x)$ in Equation 4 is calculated as $F(s_1, s_2, x) * \frac{\nu_{compartment(x)} * dt}{d_{compartment}}$ where, $\nu_{compartment(x)}$ and $d_{compartment}$ are respectively the average velocity of particles in compartment x and the distance between the mid-points of two adjacent compartment, which is the distance particles have to travel to move to the next spatial compartment. dt is the time-step. The values of various parameters used in the model are provided in Table 2.

Table 2: Parameters for PBM from Anik's hetero. agg. paper. currently place holder

Parameter	Symbol	Value	Units
Time step	δt	0.5	s
Total granulation time	T	$20,120$	s
Velocity in axial direction	v_{axial}	1	ms^{-1}
Velocity in radial direction	v_{radial}	1	ms^{-1}
Aggregation constant	β_0	1×10^{-12}	–
Initial particle diameter	R	15	μm
Breakage kernel constant	B	5×10^4	–
Diameter of impeller	D	0.1	m
Impeller rotation speed	RPM	300	rmp
Minimum granule porosity	ϵ_{min}	0.1	–
Consolidation rate	C	1×10^{-3}	–
Total starting particles in batch	$F_{initial}$	1×10^6	–
Liquid to solid ratio	L/S	0.7	–
Number of Compartments	c	3	–
Number of first solid bins	s	16	–
Number of second solid bins	ss	16	–

3.2.2. Parameters

3.3. PBM Parallel C++

3.3.1. Discretization & Parallelizing PBM

The PBM was discretized by converting each of its coordinates in to discrete bins. For the spatial coordinates a linear bin spacing was used. For the internal coordinates, solid,liquid, and gas a nonlinear binning was used. get more details from Anik on this will probably need more detail on binning for reproducability

Once the PBM had been discretized (compartmentalized/binned) a finite differences method was used which created a system of ordinary differential equations (ODEs). The numerical integration technique used to evaluate the system of ODEs was first order Euler integration check with Anik that is what we used as it is commonly used to solve these types of systems and author found it improved speeds while having minimal impact on accuracy. To obtain the most optimal parallel performance, when solving the PBM, work loads were distributed in a manner which took into account the shared memory and distributed memory aspects of the clusters the PBM was being run on. To parallelize the model in a why which could take advantage of shared memory but still effectively run across a distributed system both OMP and MPI were implemented.

One MPI process was used per CPU socket and one OMP process was used per CPU core, as authors (Bettencourt et al. (2017)) found it resulted in the best performance. MPI was used for message passing from one node to another while OMP was used for calculations on each node that could be efficiently solved using a shared memory system i.e. calculations were inter-dependent but could be computed simultaneously.

Pseudo code is presented below illustrating how the calculations are distributed and carried out during the simulation. For each time step the MPI processes are made responsible for a specific chunk of the spatial compartments. Then each OMP thread, inside of each MPI process, is allocated to one of the cores of the the multi-core CPU the MPI process is bound too. The OMP processes

divide up and compute $\Re_{agg}$ and $\Re_{brk}$. (include more detail about how they do it? last paper reviewer complained that could not understand figure by JUST reading what I wrote about it in meat of paper)

After $\Re_{agg}$ and $\Re_{brk}$ are calculated the MPI processes calculate the new PSD value for their chunk at that specific time step, $F_{t,c}$. The slave processes send their $F_{t,c}$ to the master processes which collects them into the complete $F_{t,all}$. The master process then broadcasts the $F_{t,all}$ value to all slave processes.

A crucial feature of the PBM is that the current PSD ($F_{t,all}$) value is used to compute a new time step size for the next iteration. This means all of the MPI processes need to have the same dynamic time step size at each iteration for the calculations to be properly carried out in parallel. Since the completely updated $F_{t,all}$ value is shared before calculating a new time step each process will have the same $F_{t,all}$ value. As a result each process calculates the same size for the new time step. Did not include the liquid and gas PBMs in this but hoping they will be some what assumed? Also the Ragg omp distributed work is an a what about private OMP vars specified that has impact on how model is solved etc. Should look into this. might change based on locking/blocking tests that need to be implemented still.

3.4. RP & PBM+DEM Communication

4. Results

4.1. PBM

4.1.1. PBM Validation

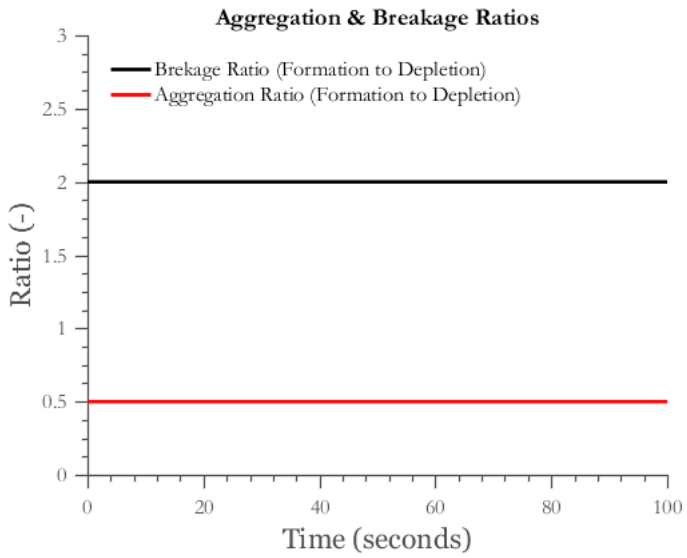


Figure 4: Ratio of formation-to-depletion through aggregation and breakage over time. Breakage ratio of 2 and aggregation ratio of 0.5 indicate mass conservation in the model. NOTE DID NOT HAVE .fig file for this figure so it is in as JPG will need to replace

The ratio between the number of particles formed due to aggregation and the number of particles depleted due to aggregation and the ratio of the number of particles formed due to breakage to the number of particles depleted due to breakage are plotted. In aggregation two particles agglomerate to form one particle and in breakage one particle breaks to form two particles. So, these ratios

Algorithm 1 Pseudo code

while $t < t_{final}$ **do**

// the spatial domain is divided into equal chunks (with in 1 bin size)

// each MPI process is assigned on chunk of spatial domain shown as c_{low} to c_{up}

// sum all c_{low_i} to c_{up_i} is = to [0,numCompartments]

for each MPI processes **do** $c = c_{low_i}$ to c_{up_i}

// each MPI process is further divided with OMP to take advantage of multi-core CPU

// each OMP process is allocated to a single compute core

// $\Re$ integrals $(i1)$ $\int_0^{s_2}$, $(i2)$ $\int_0^{s_{max_2}-s_2}$, and $(i3)$ $\int_0^{s_{max_2}-s_2}$ are divided into smaller integrals

// $\int_{i_1 low_n}^{i_1 up_n}$, $\int_{i_2 low_n}^{i_2 up_n}$, and $\int_{i_3 low_n}^{i_3 up_n}$ which are solved by the "n" OMP processes

// allocated to that MPI process (CPU)

for each OMP process **do**

$$\Re_{agg}(s_1,s_2,c) = \frac{1}{2}\int_0^{s_1}\int_{i_1 low_n}^{i_1 up_n}\beta(s_1',s_2',s_1-s_1',s_2-s_2',c)F(s_1',s_2',c)F(s_1-s_1',s_2-s_2',c)ds_1'ds_2' - F(s_1,s_2,c)\int_0^{s_{max_1}-s_1}\int_{i_2 low_n}^{i_2 up_n}\beta(s_1,s_2,s_1',s_2',c)F(s_1',s_2',c)ds_1'ds_2'$$

$$\Re_{break}(s_1,s_2,c) = \int_0^{s_{max_1}}\int_{i_3 low_n}^{i_3 up_n}K_{break}(s_1',s_2',c)F(s_1',s_2',c)ds_1'ds_2' - K_{break}(s_1,s_2,c)F(s_1,s_2,c)$$

end for

$$F_{t,c} = \frac{\Delta F(s_1,s_2,c)}{\Delta t}\Delta t + F(s_1,s_2,c)_{t-1}$$
$$= (\Re_{agg}(s_1,s_2,c) + \Re_{break}(s_1,s_2,c) + \dot{F}_{in}(s_1,s_2,c) - \dot{F}_{out}(s_1,s_2,c))\Delta t + F(s_1,s_2,c)_{t-1}$$

end for

MPI Send $F_{t,c}$ to Master MPI process

MPI Recv $F_{t,c}$ from MPI all slave processes

Master consolidate all $F_{t,c}$ chunks into a complete $F_{t,all}$

Master does inter-bin particle transfers (updates $F_{t,all}$)

MPI Bcast $F_{t,all}$ to all slave processes

$t_{new} = t + timestep$

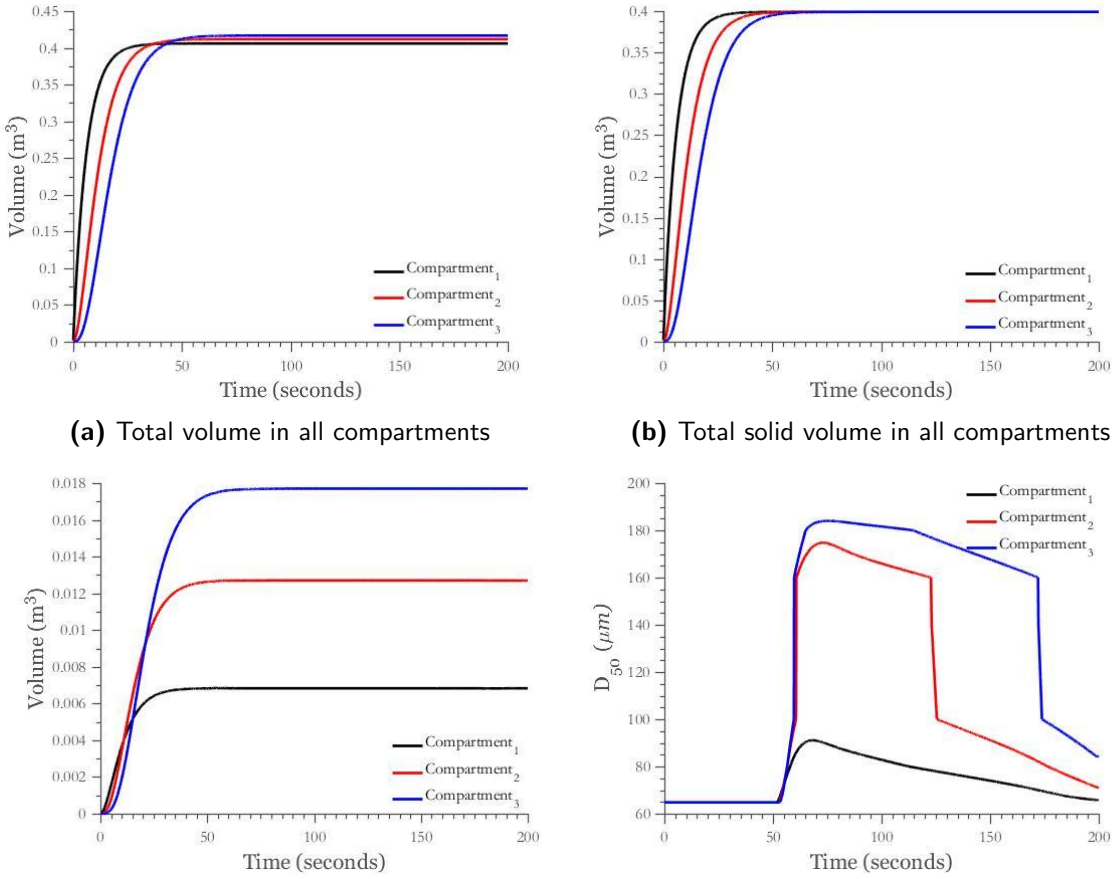
end while

are expected to be 0.5 and 2 respectively. As can be seen from Figure 4, these ratios are accurate confirming that mass is conserved accurately in the model.

The granulator was divided into 3 compartments spatially and the total volume, solid volume and pore volume and the median diameter d_50 in each compartment were plotted to study the granulation behaviour and are shown in Figure 5.

It can be seen from Figure 5a that the total volume starts to increase first in compartment 1 followed by compartment 2 and then compartment 3. This happens as gradually particles entering compartment 1 moves to the other compartment due to particle transfer from compartment 1 to compartment 2 and then compartment 3. In Figure 5b it is observed that the solid volume similar to the total volume increases first in compartment 1 and last in compartment 3. The solid volume

becomes constant and equal in all the compartments at around 30-50 seconds and steady state is reached when the rate of particle volume being transported through the compartments and leaving the system is equal to the rate of particles entering the system. Although, as seen in Figure 5c the pore volume which is the sum of the gas and the liquid volume is highest in compartment 3 and lowest in compartment 1. This happens due to the external liquid addition to the system. As the particles move from compartment 1 to compartment 3, they gradually acquire a higher amount liquid, thereby increasing the pore volume. In Figure 5d, the D_50 is seen to be increasing from compartment 1 to 3. This happens because of the size enlargement of large particles coming in from the previous compartment because of the external liquid added to each compartment and a longer residence time in the granulator.



(a) Total volume in all compartments

(b) Total solid volume in all compartments

(c) Total pore volume in all compartments

(d) D_{50} in all compartments

Figure 5: Volume and D50 in all compartments over time. Volumes become constant as steady state is reached. Median diameter increases and then decreases as bigger particles leave the system and smaller particles occupy that volume.

4.1.2. Parallel C++ PBM Validation

show PSD or D_50 is the same as Matlab or serial PBM

1. fig D_50 Matlab vs Parallel

4.1.3. Parallel PBM Performance

show that RP has minimal impact on performance

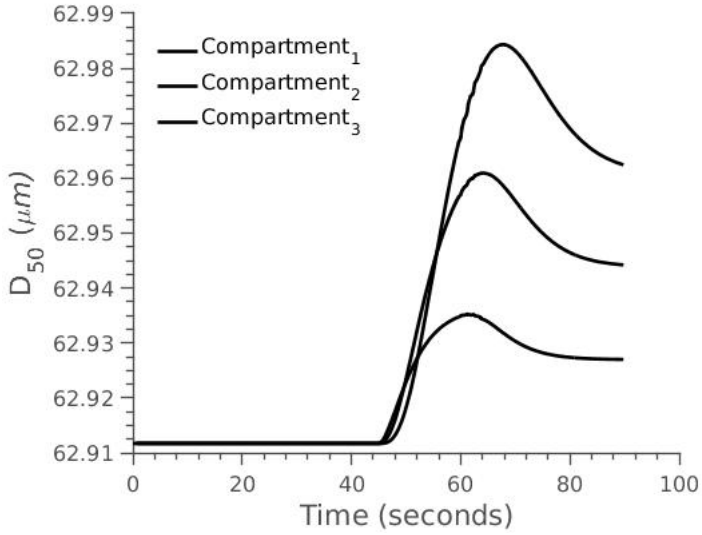


Figure 6: D_{50} of Matlab PBM vs Parallel PBM

show that performance is mostly unaffected by RP

1. fig scaling

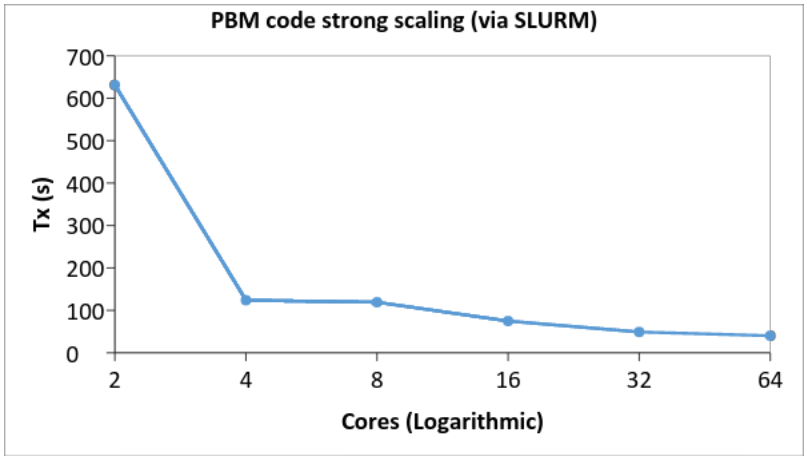


Figure 7: PBM strong scale slurm

2. fig scaling w/ RP

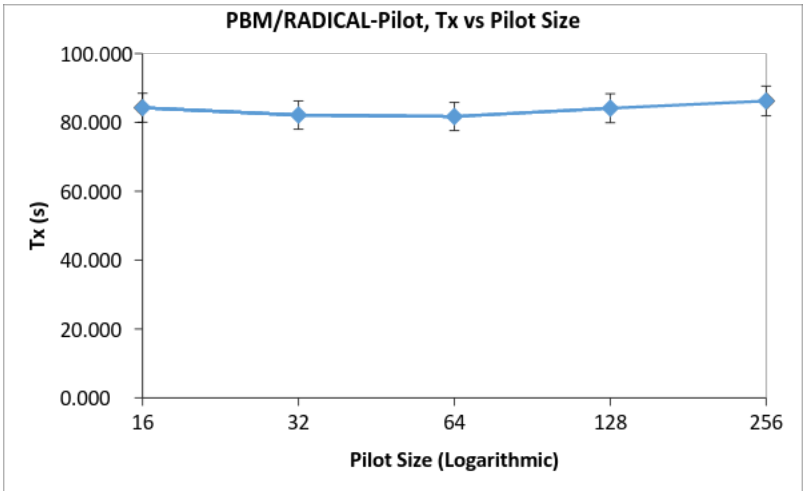


Figure 8: PBM strong scale RP

4.1.4. Parallel PBM Parameter space and Parameter Estimation

I. show how effective parallel pbm is for parameter estimation

II. Find/ explore ranges of DEM data that PBM can use to find Critical parameters and sensitivities will be useful to us in linking and in picking best DEM parameters to vary and best parameters for PBM+DEM code

1. fig range of some parameters?
2. fig range of other pbm parameter ?

4.2. DEM

4.2.1. DEM Validation and Parameter Space Studies

show that DEM is somewhat behaving like a real system

fig constant flow reached by end of DEM simulation

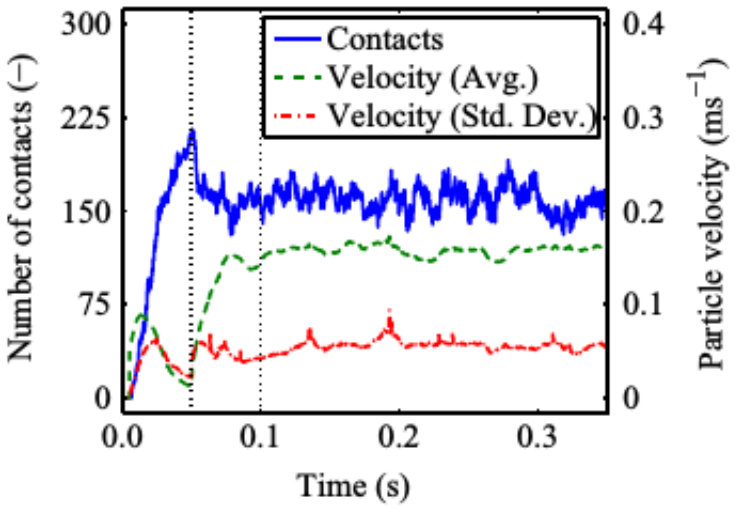


Fig. 3. Time trajectories from DEM simulation showing number of contacts between 1-mm particles with particles of the same size, and the average and standard deviation of the velocity of 1-mm particles. Particles are added at time=0 s with a median diameter of 1.5 mm and a standard deviation of 0.5 mm. Dotted lines indicate start of rotation of the screw and start of data collection, respectively.

Figure 9

fig RTD

show affects of certain parameters on DEM and which ones are most critical to outcomes - will help decide on PBM+DEM parameters to study in later section

fig vary impeller RPM see how RTD or hold up changes

fig vary PSD (range and/or particle sizes) to see how C_{coll} etc will be affected - important for PBM Kernel

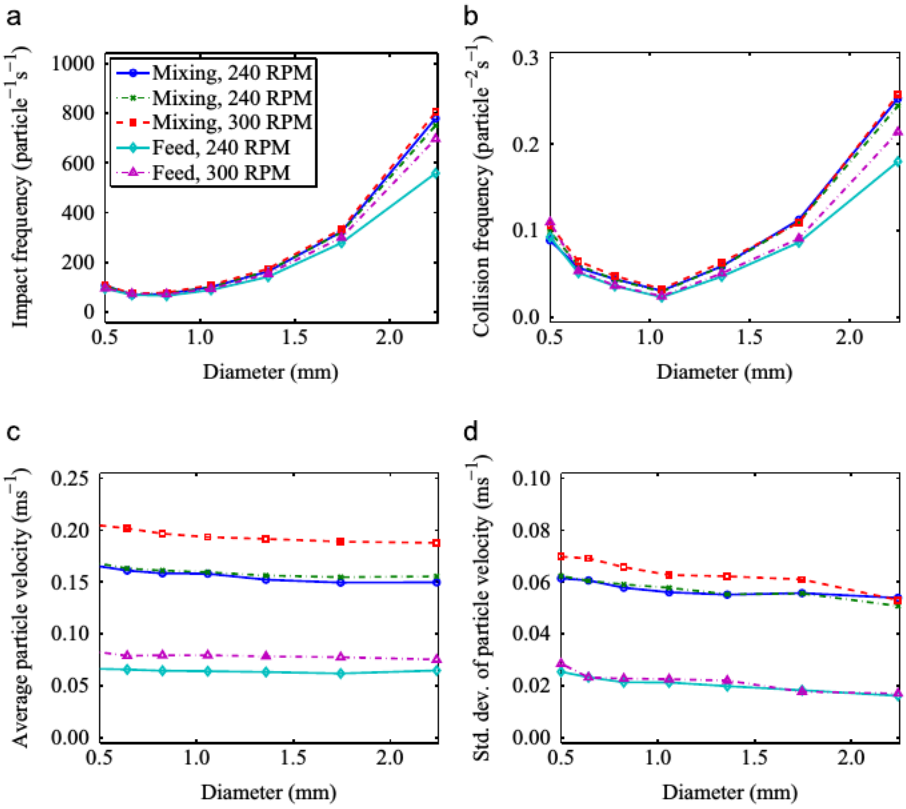


Fig. 6. DEM results showing the effects of screw element type and rotational speed on (a) impact frequency, (b) collision frequency with 1-mm particles, (c) average particle velocity, and (d) standard deviation of particle velocity for each size class.

Figure 10: fig showing sensitivity of C_{coll} and etc to RPM

453 fig vary impeller RPM see C_{coll} changes

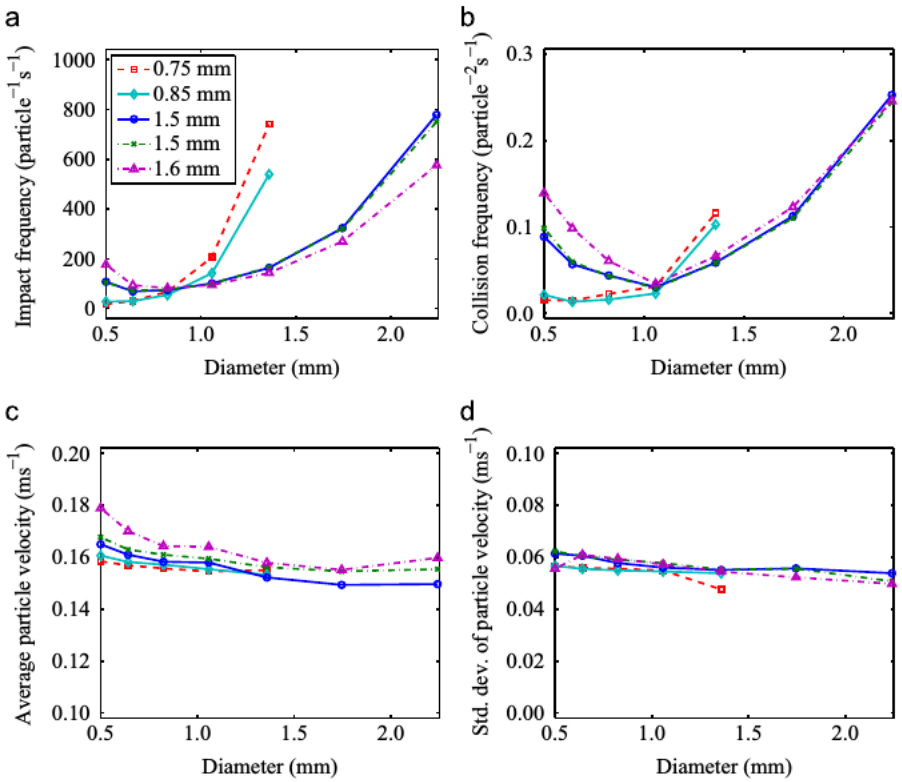


Fig. 4. DEM results showing the effects of size distribution on (a) impact frequency, (b) collision frequency with 1-mm particles, (c) average particle velocity, and (d) standard deviation of particle velocity for each size class. Median diameters are listed in the legend. The standard deviation of the diameter is fixed at 0.2 mm for the smallest two simulations and 0.5 mm for the larger simulations.

Figure 11: fig from dana 2015 mechanistic bi-directional

4.2.2. DEM Spacial Decomposition Studies

The effect of spacial decomposition on the simulation time

Speed improvements and issues using only MPI and hybrid (MPI + OpenMP)

	Total Number of Cores	Slicing in X	Slicing in Y	Slicing in Z	# OpenMP Threads	Time for 0.5 sec simulation (minutes)	Projection for 30s simulation (minutes)
MPI - only	64	64	1	1	1	10.27	**616 (10.27 hrs)**
	32	32	1	1	1	23.4	**1404 (23.4 hrs)**
Hybrid	64	8	1	1	8	10.25	**616(10.25 hrs)**
	64	16	1	1	4	8.7	**522 (8.7 hrs)**
	64	32	1	1	2	7.47	**448 (7.5 hrs)**
	64	8	2	2	2	6.83	**410 (6.8 hrs)**
	64	16	2	1	2	7.2	**430 (7.2 hrs)**
	64	4	2	2	4	7.96	**480 (8 hrs)**

Figure 12: The effect of spacial decomposition on the performance of the simulation

fig vary PSD (range and/or particle sizes) to see how C_{coll} etc will be affected - important for

458 PBM Kernel

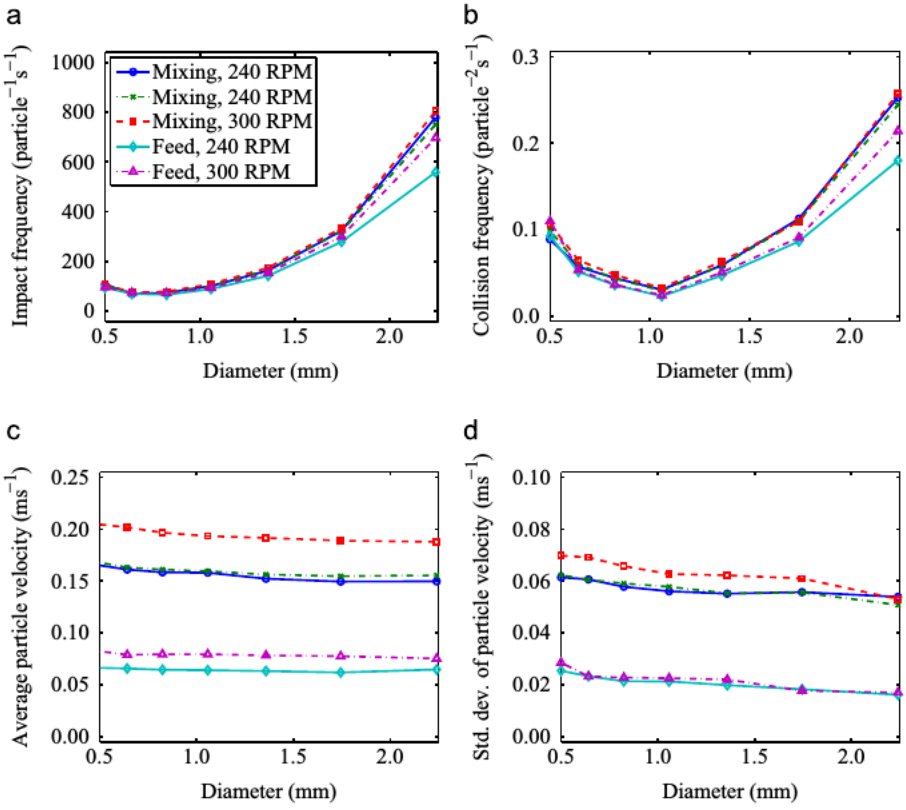


Fig. 6. DEM results showing the effects of screw element type and rotational speed on (a) impact frequency, (b) collision frequency with 1-mm particles, (c) average particle velocity, and (d) standard deviation of particle velocity for each size class.

Figure 13: fig showing sensitivity of C_{coll} and etc to RPM

459 Talk about over-slicing or excess decomposition(confirm with profs)

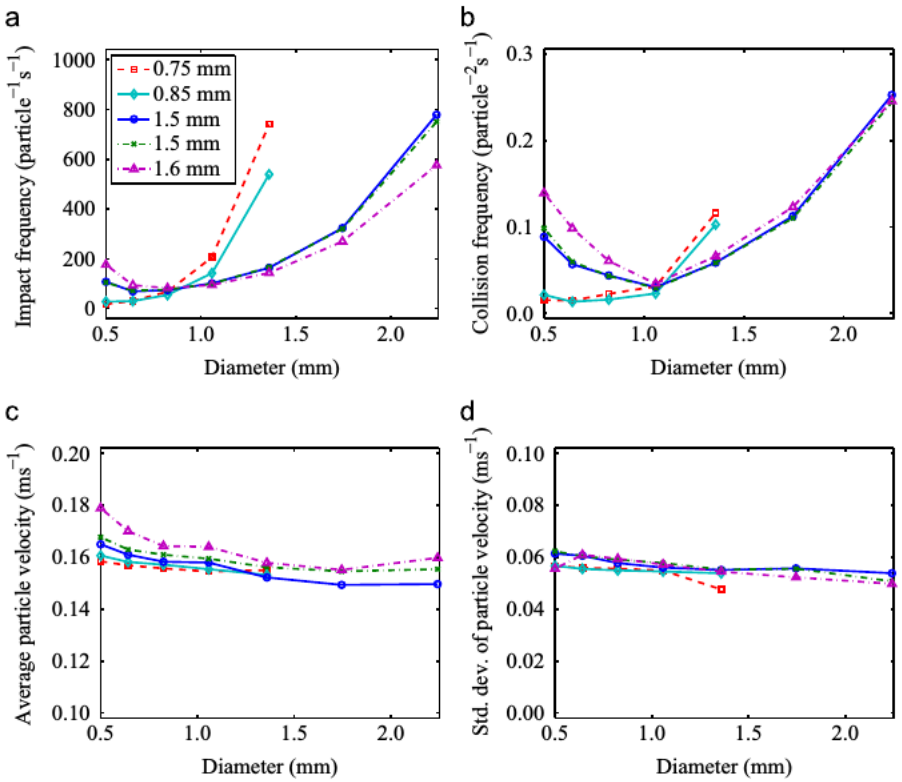


Fig. 4. DEM results showing the effects of size distribution on (a) impact frequency, (b) collision frequency with 1-mm particles, (c) average particle velocity, and (d) standard deviation of particle velocity for each size class. Median diameters are listed in the legend. The standard deviation of the diameter is fixed at 0.2 mm for the smallest two simulations and 0.5 mm for the larger simulations.

Figure 14: fig from dana 2015 mechanistic bi-directional

4.2.3. DEM Performance

Talk about how the time for simulation varies with # of cores

1. fig scaling

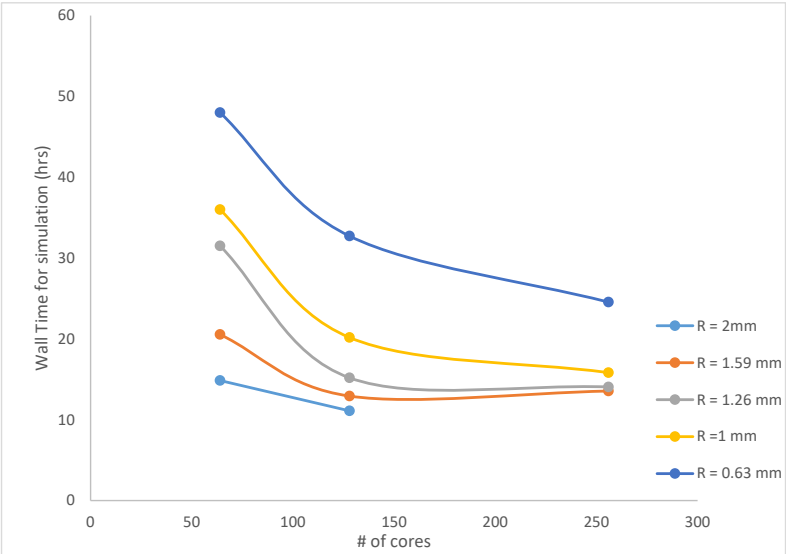


Figure 15: The variation in the amount of time taken for the simulation as a function of # of cores

2. fig scaling w/ RP

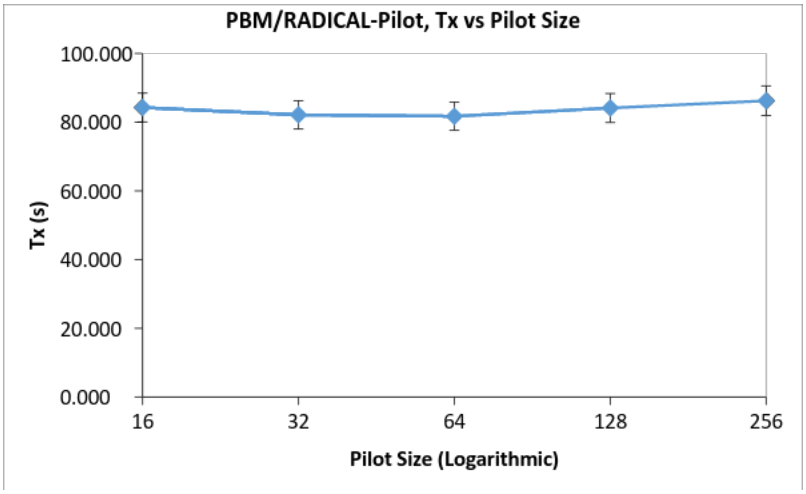


Figure 16: DEM scale with RP

4.3. PBM+DEM - RP

4.3.1. PBM+DEM Validation/Accuracy?

Talk about how the physics remain the same whether mono-sized particles are used or a size distribution is used.

Figs comparing the one-way coupling from both these sources.

4.3.2. PBM+DEM Performance

strong scaling

fig PBM + DEM RP strong scaling

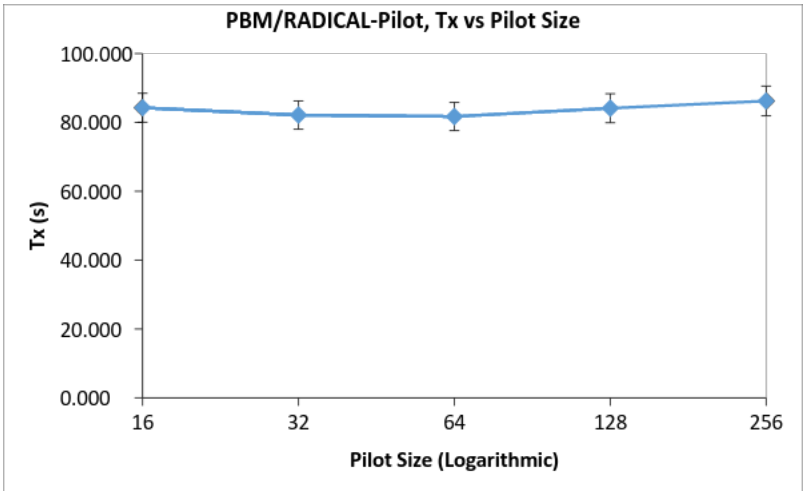


Figure 17: PBM+DEM scale with RP

4.3.3. PBM+DEM Parameter studies

show how PBM+DEM captures multi-physics as parameters changed. helps validate and support model development. show we have made a useful tool for future work.

fig

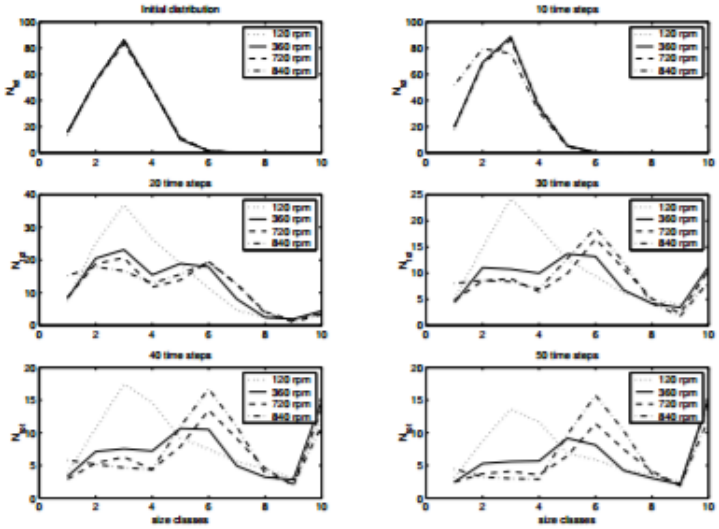


Figure 8: PSD evolution for simulations run at 120 rpm, 360 rpm, 720 and 840 rpm.

Figure 18: PBM+DEM scale with RP

5. Conclusions

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