

# PARTICLES SIZE DISTRIBUTION EFFECT ON 3D PACKING OF NANOPARTICLES INTO A BOUNDED REGION

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**Abstract** In this paper, the effects of two different Particle Size Distributions (PSD) on packing behavior of ideal rigid spherical nanoparticles using a novel packing model based on parallel algorithms have been reported. A mersenne twister algorithm was used to generate pseudorandom numbers for the particles initial coordinates. Also, for this purpose a nanosized tetragonal confined container with a square floor (300 \* 300 nm) were used in this work. The Andreasen and the Log-normal PSDs were chosen to investigate the packing behavior in a 3D bounded region. The effects of particle numbers on packing behavior of these two PSDs have been investigated. Also the reproducibility and the distribution of packing factor of these PSDs were compared.

**Keywords** Random Packing; Particle Size Distributions; Andreasen

**چکیده** در این مقاله تاثیر دو توزیع اندازه ذرات بر روی رفتار تراکمی نانوذرات کروی صلب ایده آل توسط یک مدل تراکم با الگوریتم موازی گزارش شده است. الگوریتم مرسن توئیستر به منظور تولید اعداد شبه تصادفی برای مختصات اولیه ذرات بکار گرفته شده است. بدین منظور از یک محفظه مکعب مستطیلی شکل با قاعده مربعی (300×300 nm) استفاده شد. توزیع اندازه ذرات آندریازن و لگاریتم - نرمال به منظور بررسی رفتار تراکمی سه بعدی ذرات در یک ناحیه محدود بکار گرفته شده است. تاثیر تعداد ذرات بر رفتار تراکمی این دو توزیع اندازه ذرات بطور گسترده بررسی شده و نیز تکرارپذیری و توزیع ضریب تراکم در این توزیع ذرات مقایسه شده است.

## 1. INTRODUCTION

Random packing of particles has been studied for many years because of their interesting geometrical properties and technological applications in porous media [1]. The term of particle 'Packing' means 'putting together' and 'arranging' particles in a confined space. Random packing means that all particles of the same size and shape have the same probability to occupy each unit volume of a

mixture [2]. Particle packing in ceramic processing has a significant effect on shrinkage and density of bodies during sintering as well as the properties of the final products. It also affects the efficiency of powders during compaction and the final surface properties and corrosion behavior of powder metallurgy products [3-9].

Research on particle packing can be classified into three approaches: experimental, theoretical, and numerical (computer simulation). Due to

difficulties associated with experimental methods of investigating packing behavior theoretical and computational approaches are usually preferred [10].

Many parameters affect the packing characteristics of particles, such as particle size distribution (PSD), particle shape, interparticle friction, elasticity, surface chemistry (interparticle forces), agglomeration and vibration (rearrangement of particles upon shaking) [11-20]. Among these parameters, due to its ease of adjustment in technical applications particle size distribution is probably the most significant.

Random sphere packing has been used as porous media models to simulate pore-scale phenomena such as drainage and imbibition, dispersion, sorption and diffusion, flow through porous media and transport properties [21,22].

Most of the algorithms for random packing of spheres fall into two major types of models: sequential addition and collective rearrangement models. In the sequential addition models, new particles are added to the existing system. Subsequently, the newly added particles are moved to a stable place that all momenta made by the gravity and the supports satisfy each other [23]. In the collective rearrangement models, the particles coordination are randomly generated and they are allowed to overlap. After this stage, the relaxation process is done to separate overlapped spheres to reach the stable position [24].

The former may result in lower packing factors in comparison with experimental values while the latter method gives a non-realistic network of contacts [25].

A random variable  $x$  is said to be Log-normally distributed if  $\log(x)$  is normally distributed. Only positive values are possible for the variables, and the distribution is skewed to the left. Two parameters are needed to specify a Log-normal distribution. Traditionally, the mean  $\mu$  and the standard deviation  $\sigma$  (or the variance  $\sigma^2$ ) of  $\log(x)$  are used. However, there are clear advantages in using "back-transformed" values (the values are in terms of  $x$ , the measured data):

$$\mu^* = e^\mu \quad (1)$$

$$\sigma^* = e^\sigma \quad (2)$$

$X \sim \Lambda(\mu^*, \sigma^*)$  is then used as a mathematical

expression meaning that  $X$  is distributed according to the Log-normal law with median  $\mu^*$  and multiplicative standard deviation  $\sigma^*$ . The median of this Log-normal distribution is  $\text{med}(X) = \mu^* = e^\mu$ , since  $\mu$  is the median of  $\log(X)$ . The dispersions produced by milling, grinding or crushing provide examples of powders with Log-normal distribution [26].

In the late 1920s Andreasen and Andersen developed a continuous particle size distribution for the particles packing [27]. Although Andreasen's work had not received much attention when it was published, it has gained increasing recognition recently [28].

Packed particles can be unconfined where there are no containing walls or when particles can be packed in a wide variety of structural containers. The bounding region affects the packing properties of particles that are important in the investigation of nano-sized systems. In the case of random packs, a so-called wall effect exists because the proximity of a solid surface will introducing some local order into a random packing. Thus, the particles next to the solid surface tend to form a layer of the same shape as the surface. This so-called base layer is a mixture of a cluster of square and triangular units. Randomness increases with increasing distance from the base layer, with resultant disappearance of the distinct layer. Another important aspect of wall effects is the existence of a region of relatively high void age next to the wall due to the discrepancy between the radii of curvature of the wall and the particles [29]. Also no rearrangement was done to the particles in their stable state.

## 2. MODELING

We assumed particles to be ideal rigid hard sphere with two different particles size distribution i.e. Andreasen [30] Equation 3 and a typical Log-normal [31] Equation 4 with equivalent parameters. Thus the particles are designated by the coordinates of their centers and radii.

$$f(D) = \frac{n}{D} \left( \frac{D}{D_{\max}} \right)^n \quad n = 0.5 \quad (3)$$

Where  $D$  is particle diameter,  $D_{\max}$  is the largest

particle size and  $n$  is the distribution modulus.

$$f(D) = \frac{1}{D \cdot \sigma \sqrt{2\pi}} \exp\left(-\frac{1}{2\sigma^2} (\log(D) - \mu)^2\right) \quad (4)$$

In this work, it is assumed that the friction is infinite. This assumption affects the obtained values for the packing factors but doesn't affect the behavior of the system from a standpoint of reproducibility, also the assumption affects both PSD's packing factor values so their reproducibility can be compared [32,33].

A simple Cartesian coordinate system is applied to all of the computations inside the simulation environment. Coordinates in the system are referred to as either X, Y and Z. The environment is oriented so that the Z-axis is in the direction of the gravitation field and the origin set in the corner of the container in the simulation process.

A boundary region was a tetragonal confined container with a square floor. The bottom of the container is flat and located on the X-Y plane, as shown in Figure 1. All the locations, movements and forces are described and stored in the coordinate system through the simulation. As particles exhibit spherical symmetry, it's sometimes useful to do computations on a particle using spherical coordinates, (where the origin is the center of the mass). For these cases all vectors have been described in the spherical coordinates:  $\alpha$ ,  $\theta$  and  $r$ , where  $\alpha$  describes angle on the X-Y

plane (between 0 and  $2\pi$ ),  $\theta$  describes angle on the vector and the Z axis (between 0 and  $\pi$ ) and  $r$  is the length of the vector. This coordinate system was used to estimate the stability of particles in the presence of three or more contact points.

Walls of the packing space (container) are described in a similar way to the particles. The most significant difference is in considering walls to be unmovable.

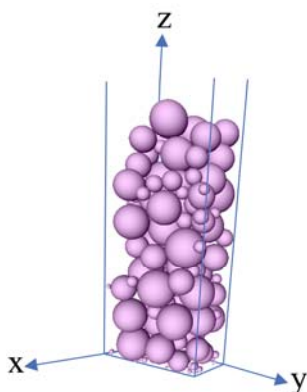
### 3. PACKING ALGORITHM

A novel time-independent model for the packing of particles based on the Event Dynamics (ED) model has been created. The collisions were considered to be soft. This naturally requires the simulation to compute the effects several times during the collision. This causes the model to require more computational power than conventional ED model but in the case of no collision occurrence, the present model works much faster.

The particles' X-Y coordinates were randomly generated. The particle then drops until it reaches the floor ( $Z = 0$ ) or another particle. The falling of particles into the container was in random sequence. The particle's height (Z) found from the statical stability of the particles on each other. The resulting state may be described as random loose packing [34].

The stability of particles calculated based on the location of the particles existing in the container and the next movement of the particles in each calculation step determined from particle's coordinate and its contact points with boundary walls or other particles.

If the falling particle contacts with the existing particles in the container, the dropping particle will roll around the standing particle towards the ground. If a second particle is encountered in the process, the dropping particle will roll around the first and second standing particles until a third or more contacts (the floor, another particle, or a wall) when the momenta about each contact point is computed and if they satisfy each other, the particle is reached to its stable position. This deposition process is deterministic, after starting the location and particle size are randomly generated.



**Figure 1.** 3D image of a typical Andreasen packing.

#### 4. PARALLEL ALGORITHM AND IMPLEMENTATION

The present parallel algorithm for computing packing factors uses an implicit parallelism paradigm. In every stage of parallelism each task is the result of the partitioning algorithm invoked at the first iteration of the program that uses functional decomposition. This partitioning and using implicit parallelism leads this researcher to the point where it can send these tasks to each node in the cluster. Also, load balancing is achieved by the sending of the next tasks who send their result surely if the checking algorithm finds that the data sent by the node is incomprehensive it sends again the task to another node and if the node gives the standard result the data will be accepted and the node which has given the wrong result will be given a negative point. If the node reaches a threshold it will be disbanded by the server and its tasks will be given to other nodes. The tasks which will be executed on the node have the same order of computation and will be finished simultaneously. If the other that has been given the task sends the wrong result again that will mean that the task has a problem so it will be registered on a file with initiation and will be checked by hand on another system to find the problem. After discarding and registering the tasks the server will resume its work on other tasks.

The parallel algorithm has been implemented using the MPI library and MPICH2 implementation. This implementation enabled the researcher to use Visual Studio .NET 2003 to produce more high quality codes that are optimized for systems with Microsoft Windows XP service pack 2 operating system. This MPI implementation has an advantage over other system PVM which does not completely support object orientation. Complete object orientation was tried in the use of the present code so as to utilize the said code for future simulations. The result was a higher order of compatibility with different softwares and so the researcher was able to feed the wanted results to other simulation tools with adding a suitable formatting package to the said code and produce suitable result.

The hardware used in the simulation consisted of six dual processor PCs with 512 MB RAM which one of them was used for replication of data. These PCs were connected together in a star topology. Each simulation took almost 72 hours and the data

was gathered in the server.

#### 5. PACKING FACTOR

After finalizing the packing of particles in the container, to calculate the amount of packing factor, the normal height of box that has the average planar density of particles at the top height plane was found. The average particles planar density starting from bottom to half of the maximum particle's height was then calculated.

The packing factor of the particles obtained from the division of total volume of particles involved in the normal box with its volume.

#### 6. RANDOM NUMBERS

The present method for generating random numbers in the Mersenne twister complies with MT19937 standard which is 32-bit length number generator. This algorithm produces pseudorandom numbers which has a very long period length. Its main advantage is that it will be produced very rapidly due to use of bitwise operators and the algorithm. Although the production is fast, the produced numbers are of very high qualitative and due to this long period it can be used in highly repeated algorithms which need a lot of random numbers in each repetition. The other advantage of this algorithm is the higher order of dimensional equi-distribution comparing to the other algorithms and passing diehard tests [35,36]. In each case, four random seeds for Mersenne twister algorithm were used.

#### 7. CALCULATION PARAMETERS

The accuracy of all calculation was 0.0001 nm. The areas under the PSD diagrams were assumed to be 500 (i.e. equal to 500 particles) for 800 samples and various numbers of particles (i.e. 100, 200, 300, 400 and 500) was used to simulate 50 samples. Distribution modulus of Andreasen PSD was assumed to be 0.5. The integration of the PSD

**TABLE 1. Particles Number Resulted from Integration of PSDs.**

Particle Size Distribution	Initial Numbers of Particles	Truncated Number of Particles
Log-Normal	100	62
Log-Normal	200	157
Log-Normal	300	247
Log-Normal	400	341
Log-Normal	500	432
Andreasen	100	59
Andreasen	200	149
Andreasen	300	241
Andreasen	400	332
Andreasen	500	426

diagrams were done by 1 nm steps in lower-Riemann method. The area of the bottom plane of box was 300×300 nm. Minimum and maximum diameters of particles were 1 nm and 60 nm, respectively. The median of Log-normal PSD was assumed to be 30 nm.

## 8. RESULTS AND DISCUSSION

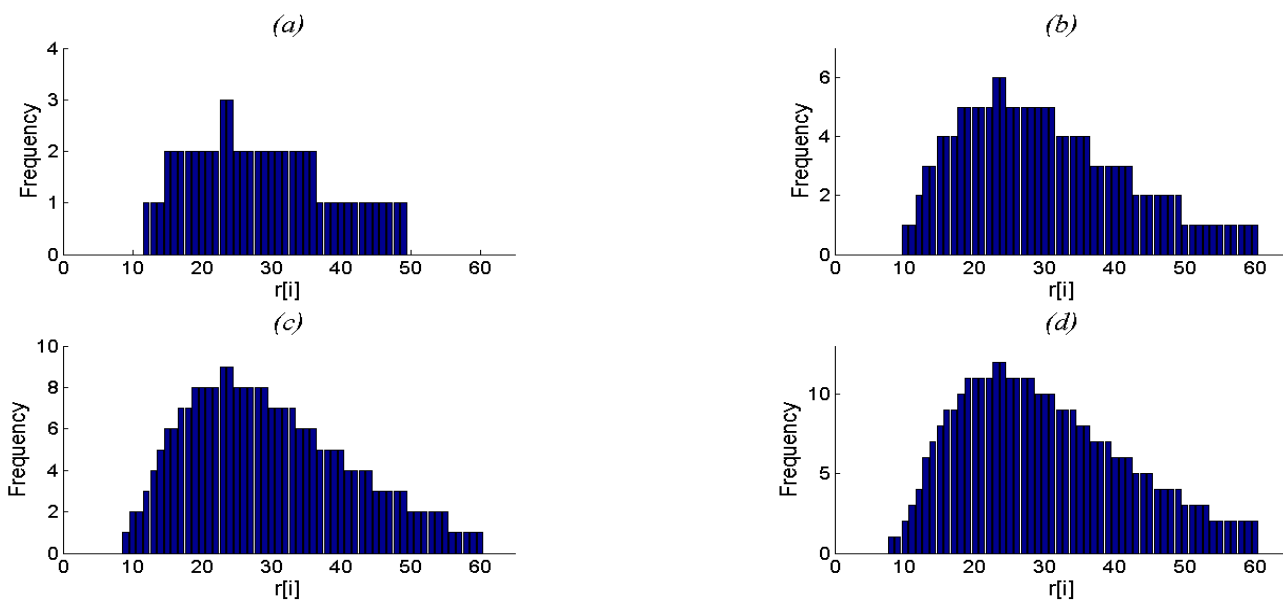
Riemann Integration of Particle size distribution and its truncation cause inaccuracy in the calculation of particle numbers. Table 1 lists the total particle numbers resulted from integration of PSDs. Also, the particle size distribution of Log- normal and Andreasen are shown in Figures 2 and 3, respectively.

In general, increase in friction coefficients of particle-particle and particle-wall contact points results in decreasing of packing factor. This is the

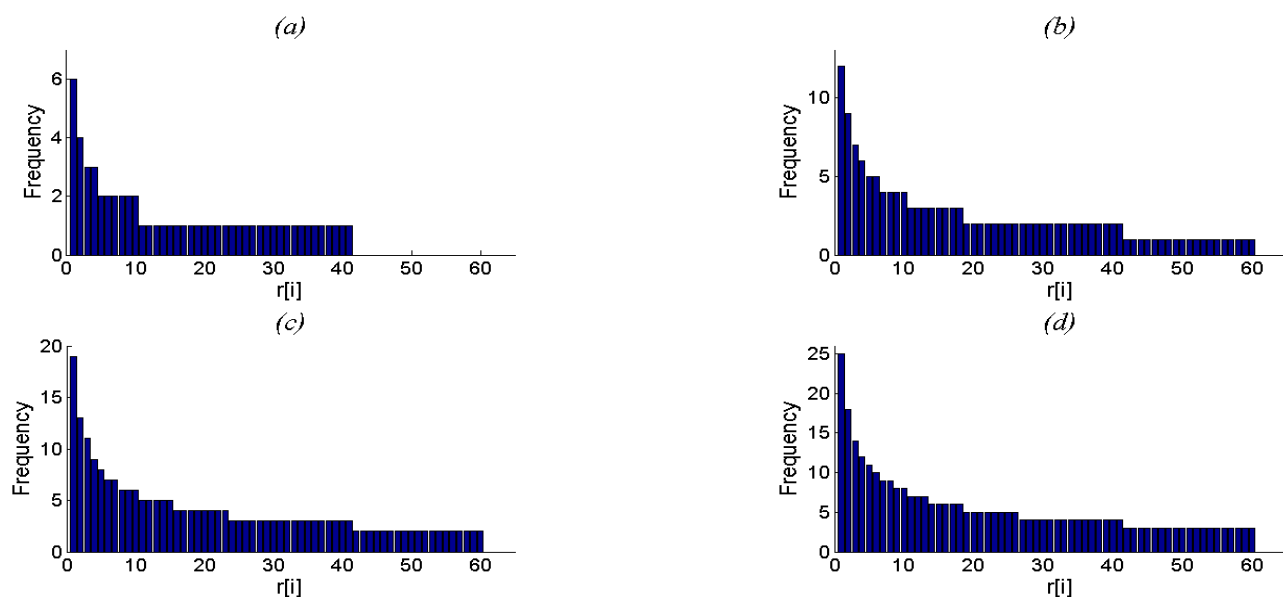
main reason why the present simulations generated relatively low packing density. Furthermore, small size of boundary region in relation to particles size imposed spatial blockage for further densification due to introducing some local order into random packing.

**8.1. Log-Normal** Figure 4 illustrates the histograms of obtained packing factor values for the 100, 200, 300, 400 and 500 particles, respectively in 50 packing simulation processes. The behavior of the packing factor by increasing of particle numbers is represented in Figure 5.

Increasing the particles number will result in decreasing the walls area to particles number ratio. As it can be seen in Figure 4, by increasing the particle numbers, the packing factor increased. This phenomenon can be the result of the decreasing wall effect. Also, by increasing particle numbers the numbers of smaller particles increase and because of their ability to



**Figure 2.** Particle size distributions of (a) 100, (b) 200, (c) 300 and (d) 400 particles in Log-normal distribution.

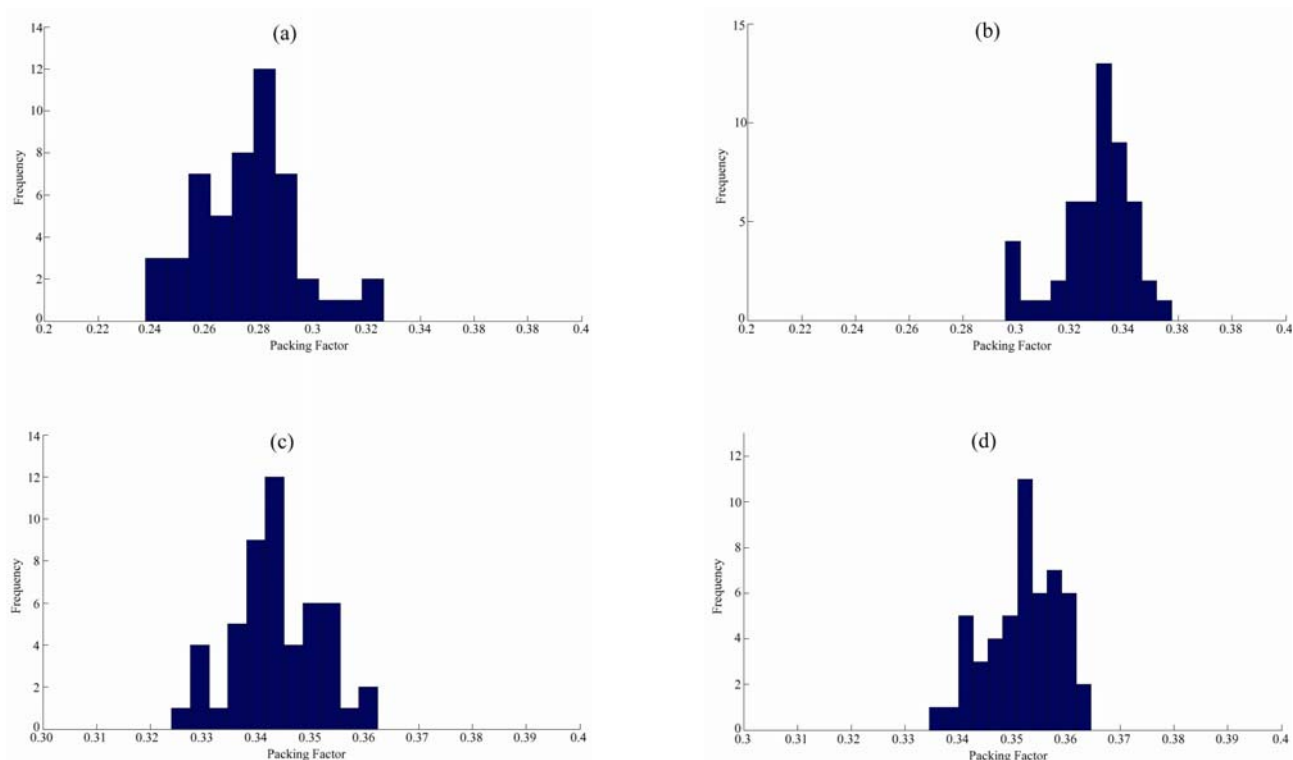


**Figure 3.** size distributions of (a) 100, (b) 200, (c) 300 and (d) 400 particles in Andreasen distribution.

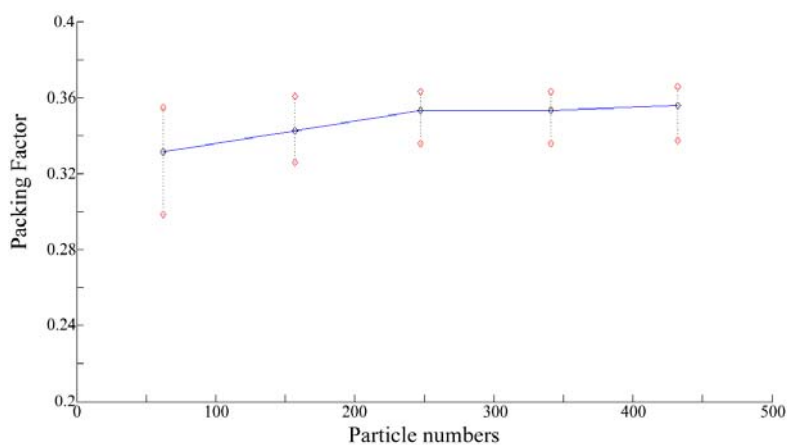
fill the voids, the packing factor was increased as indicated in Figure 5.

The decreasing of packing factor ranges is

obvious in Figure 5. The explanation of this phenomenon is that there is an increasing of particle numbers resulting in decreasing voids



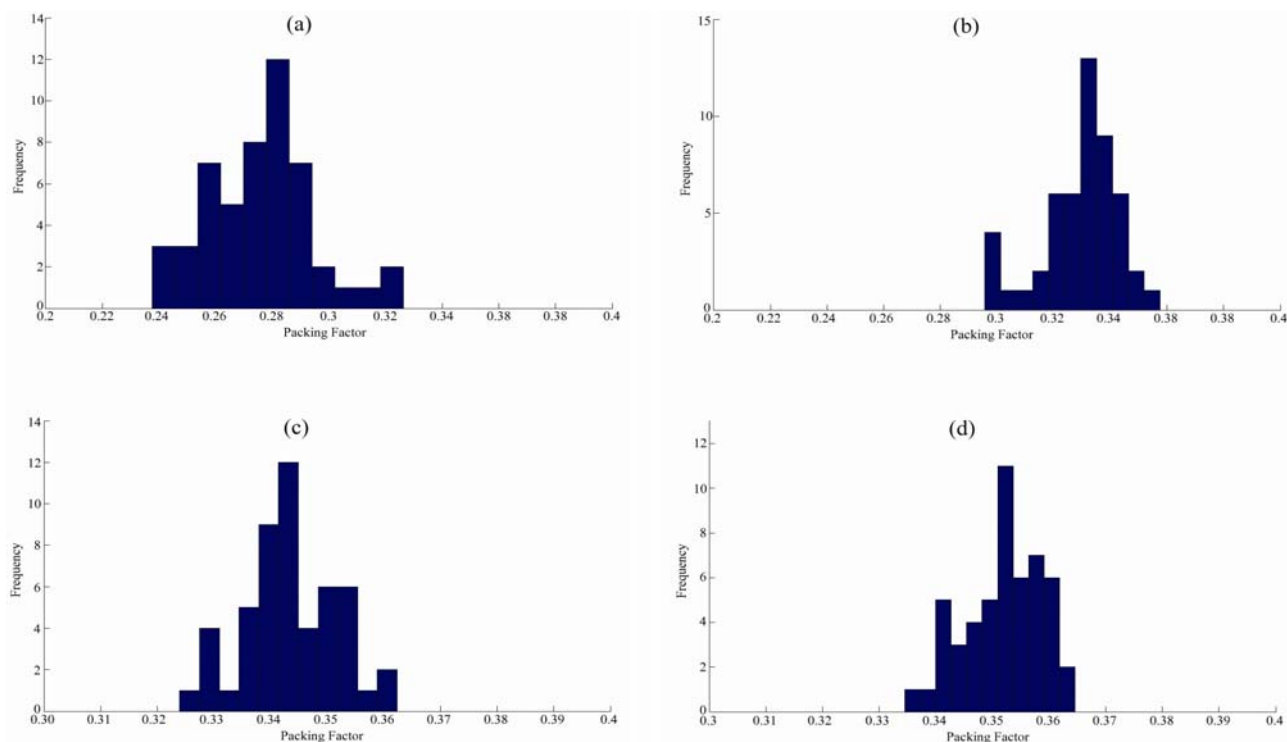
**Figure 4.** Histograms of obtained packing factor values for the (a) 100, (b) 200, (c) 300 and (d) 400 samples in Log-normal Particle Size Distribution.



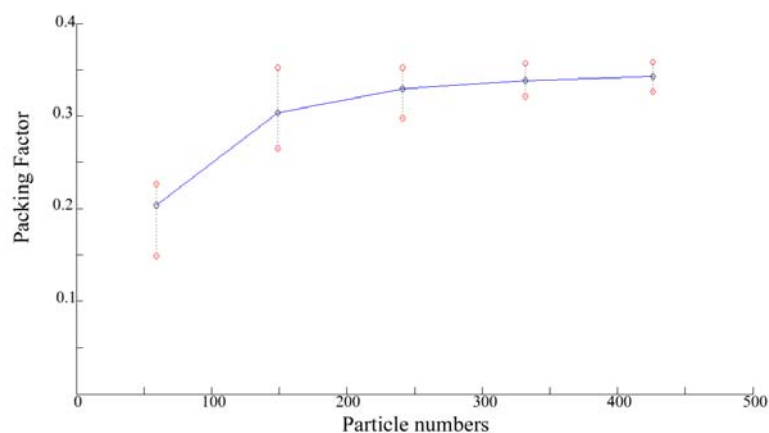
**Figure 5.** The effect of particles number on the packing factor distribution of Log-normal Particle Size Distribution. The packing factor ranges are indicated as vertical bars in the figure. The curve maps to the median values of packing factor distributions.

fraction. Thus, the range of packing factor distribution gets narrower.

**8.2. Andreasen** Figure 6 illustrates the histograms of obtained packing factor values for the 100, 200,



**Figure 6.** Histograms of obtained packing factor values for the (a) 100, (b) 200, (c) 300 and (d) 400 samples in Andreassen Particle Size Distribution.

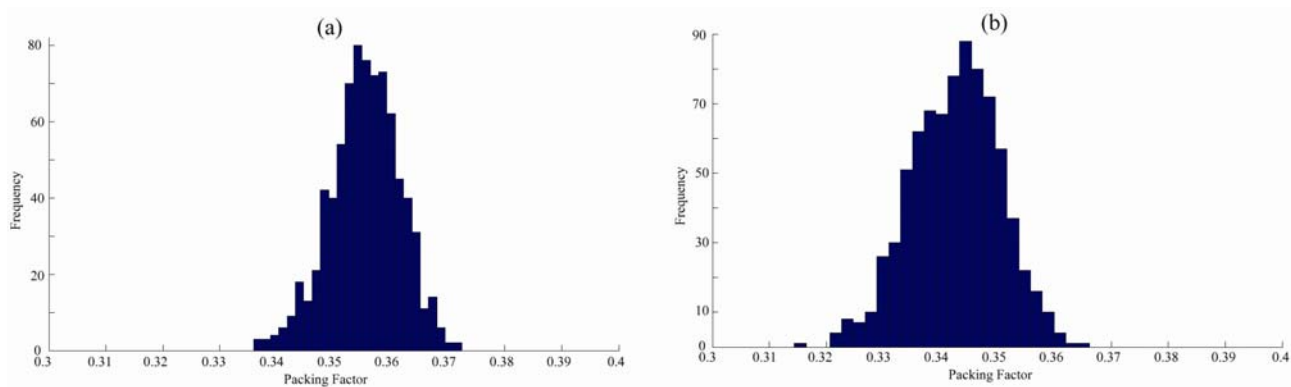


**Figure 7.** The effect of particles number on the packing factor distribution of Andreassen Particle Size Distribution. The packing factor ranges are indicated as vertical bars in the figure. The curve maps to the median values of packing factor distributions.

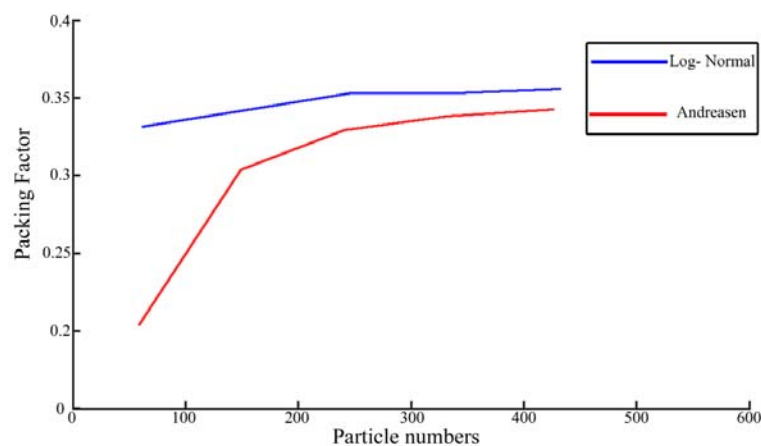
300, 400 and 500 particles, respectively in 50 packing simulation processes. The behavior of the packing factor by increasing particle numbers is represented in Figure 7.

As cited in former sections, increasing the particles number will result in a decrease of wall area to particles number ratio. Hence, by increasing the particle numbers, the Andreassen PSD packing





**Figure 8.** Histograms of obtained packing factor values for the (a) Log-normal, (b) Andreasen particle Size Distribution for 500 particles in 800 packing simulation processes.



**Figure 9.** Comparison between Log-normal and Andreasen particle Size Distribution with increasing particles numbers.

factor also increased (Figure 6).

The decreasing of packing factor ranges is obvious in Figure 7. The explanation of this phenomenon is the same as log-normal PSD with this difference that the more small size particles in Andreasen PSD exist.

**8.3. Reproducibility** Figure 8 illustrates the histograms of obtained packing factor values for 500 particles, respectively for Log-normal and Andreasen Particle Size Distributions in 800 packing simulation processes. This procedure

was to investigate the effect of particles size distribution on 3D packing of particles' reproducibility.

As can be seen in Figure 8, the packing factor of Andreasen PSD shows wider distribution range in comparison with Log-normal PSD. There are two possible sites for small particles. They can fill the voids larger than their radii or behave as a support for the subsequent bigger particles. This procedure can produce samples with various packing factors. This mechanism leads to lower reproducibility in Andreasen PSD in comparison

with Log-normal.

**8.4. Comparison** As can be seen in Figures 2 and 3, by increasing particle numbers, the proportion of smaller particles increases, but this increase in Andreasen is more than Log-normal PSD. According to the explained mechanism of smaller particles behavior, filling of voids in Andreasen PSD is occurred more than Log-normal. This phenomenon causes a tangent slope of the particle numbers vs. packing factor curve to increase (Figure 9).

## 9. CONCLUSION

A novel model for simulating random packing of particles has been developed. The model was used to generate packing systems based on Andreasen and Log-normal PSDs. The results implicate an increase of packing factor by increasing particle numbers in both PSDs. With increasing particle numbers, next to the decreasing in packing factor ranges, in both Andreasen and Log-normal PSDs; the packing factor of Andreasen PSD grows more rapidly than Log-normal. Also the packing factor of Andreasen PSD shows wider distribution range in comparison with Log-normal PSD.

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