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Particle Size Distribution for Refractory Castables: A Review

ABSTRACT

Particle size distribution is important for the development of both shaped and unshaped refractories. But it is more critical for the castables as they need to satisfy two near contradictory properties, flow and compaction (strength). Continuous particle size distribution of ordered particle sizing systems is employed primarily in advanced castables due to the many advantages such as reducing the water demand, imparting desired rheological characteristics (such as vibratory/thixotropic flow or free flow/self-leveling behavior), minimizing porosity, and maximizing particle contact for enhanced bonding and optimum strength develop-

ment. Casting and placing properties of castables, which are dependent on flow behavior, are the most critical ones as they affect refractory property development in castables. Many researchers have worked on particle size distribution for castables. Primarily the works of Furnas, Andreasen and Dinger & Funk are well accepted in the castable industries mainly because of their simple approach. This paper reviews these distribution models for castables and their suitability in practical castable development.

1. Introduction

The demand for very high quality steel by the steel-using industries has forced the iron and steel industry into several revolutionary changes over the last few decades. This had led the industry to adopt various sophisticated processes to improve the quality and productivity of steel. The changes that have occurred in steel-manufacturing processes over the last two to three decades are phenomenal and so refractory materials face severe challenges in meeting critical operational parameters at high temperatures. In addition, steel manufacturers require that refractories perform better with less down time. To meet these challenges, the attention of refractory researchers, manufacturers, and users has shifted towards unshaped refractories to substitute conventionally shaped ones. Among the various classes of unshaped refractories, castables lead in all areas of research, development, manufacturing, and application due to several inherent advantages.

Though the first castable patent was in 1923, real progress in castable technology began only after the development and commercial success of calcium aluminate cement. Refractory castables have been upgraded from simple mixes of different fractions of alumina or alumina-silicates and binder to complex and technical formulations containing various additives, suitable for different critical applications with tailored properties. Castables may be described as a blended mixture of different fractions of aggregates with bonding agent and additives, supplied in dried condition as loose powder. This dry mixture of particles is further mixed with water (or other liquid binders) at the user industry. Next, the mixture is vibrated, poured, pumped, or pneumatically shot into the application site to form the desired shape or structure which becomes rigid due to the setting of the bonding material and then fired to attain sintering and strength.

Now to make any refractory we need the constituent material in various sizes to enable a better packed and dense condition. Each size represents a specific average dimension of a particle with closed ended maximum and minimum sizes. A particle is a very small piece or part (tiny portion) of a material which can be ascribed several physical / chemical properties. Now defining the size of the particle is important.

For regular shapes, like spheres or cubes, the size can be defined by its diameter or sides, respectively. But for an irregularly shaped particle, which is the actual condition in practical cases, defining the average size is complicated. Figure 1 shows and Table 1 describes the different ways to represent the particle size of an irregularly shaped particle equivalent to the diameter of a sphere. However, conventionally for refractories, the diameter of an equivalent sieve size is used to represent the particle size in practical applications.

Now, for shaped refractories, particle size distribution is planned in such a way as to achieve maximum packing density. Hence, minimum void space will remain within the structure, resulting in greater densification and strength. This densely packed condition also remains valid even after firing at elevated temperatures for the highly compact mass as sintering is higher for such a compact structure due to a reduced diffusion path and less void space. Finally, this results in a refractory with high density, strength, resistances against corrosion and abrasion, etc. However, to achieve the highest packing, one single size (monodispersion) will never work. A monodispersion is a system in which all particles are exactly the same size.

To attain dense particle packing, one has to pack the coarse-sized particles as densely as possible. Then, without disturbing the densest packing of the coarse particles, pack the pores with medium size particles. Further, without disturbing the densest packing of the coarse and medium particles, place the fines into the pores present between the medium and coarse particles. Thus, the densest condition will be attained using three different size fractions of particles. Note that all particle sizes present within the whole range from coarsest to finest are not represented, but only a few tightly defined, discrete particle sizes, are used. Such particle size packing is called discrete packing. The discrete theory, however, is based on true monodispersion of each specifically sized group of particles where the diameter ratio between adjacent size classes must be large, say 100 (greater is better). Also, this packing concept does not allow one to move or stir the particle system once it has been packed. Discrete packing reduces the water demand and fired porosity value due to greater packing (larger particle contacts and less voids) and is also reported to impart good thermal shock resistance [1].

But for castables the requirements are different. Castables primarily need to flow and also require high density and strength, both at ambient and elevated temperatures. It is a unique requirement compared to

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Table 1 · Description of diameters of an equivalent sized sphere for an irregularly shaped particle

d_{\max}	Diameter of the sphere same to the maximum dimension of the particle
d_{\min}	Diameter of the sphere same to the minimum dimension of the particle
d_w	Diameter of the sphere having the same weight as that of the particle
d_v	Diameter of the sphere having the same volume as that of the particle
d_s	Diameter of the sphere having the same surface area that of the particle
d_{sieve}	Diameter of the sphere passing through the same sieve aperture as the particle passes through
d_{sed}	Diameter of the sphere having the same sedimentation rate as that of the particle

that of the shaped refractory and also contradictory in nature as strength develops from compaction which restricts movement due to friction. Flowability of the castables is very important for application purposes and their final performance. Better flow of any castable allows one to make a good lining and makes it possible to cast any intricate shape. In this regard, self-flowing castables are the best as no external energy is required for its movement. It can flow under its own weight and the best lining performances may be expected from these self-flowing castables. Again this flowability is primarily dependent on the particle size distribution and packing of the castable system.

So for castables, a different packing model is required compared to the packing of the shaped refractories. Castables have a continuous matrix phase (containing fine aggregates, bonding material, and additives) and no continuous aggregate phase (as in shaped refractories). Aggregates are separated from one another by the matrix phase. Hence, the close contact between the aggregates is not there. Less friction between the coarse aggregate particles and also within the whole castable mass allows them to move and flow easily. Hence for castables, the closest packing of the particles is not desirable as it restricts flow. So, a new particle size distribution concept is required, which is called continuous particle size distribution. In this distribution, particle sizes are present in a continuous manner, not in a discrete manner. Different particle size fractions are used for such a pattern. Each size fraction used for this type of distribution has a maximum and minimum size value. Where each maximum size of one particular fraction is the minimum size of the earlier coarser size fraction and the minimum fraction is the coarse fraction of the next finer fraction. Thus, the whole range of particles is present within the system in a continuous manner.

In continuous particle size distribution, the aggregate fractions are used in a relatively large number of closely sized screened fractions to fill out a continuous distribution curve [2]. These size fractions represent all the sizes in the size range while discrete distribution consists of monodispersions of particles representing only two to three fractions of the sizes in the size range. Continuous particle size distribution results in very good rheology / flow characteristics of the system at relatively low water contents with good compaction, low shrinkage, and high strength values [3].

Developmental studies on continuous particle distribution have mostly been carried out for the construction (concrete) industries and because of the similarity in property requirements of the castables the same concepts are equally important and applicable for the castables. As the ceramic particles of different sizes are not exactly similar in terms of features, some assumptions are generally considered and accepted for the distribution studies. The common assumptions are:

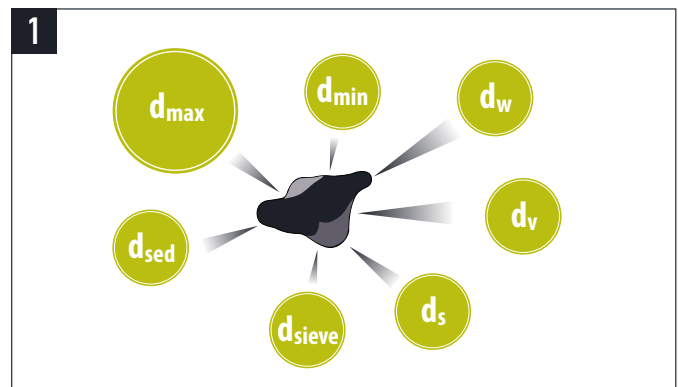


Fig. 1 • Representation of diameter of an equivalent-sized sphere for an irregularly shaped particle

- If the particles of different sizes are fully dense, hard and rigid, incompressible and uncrushable during any movement.
- The shape and size distribution of the particles present in each grain size (range) are uniform and there will be no segregation of particles on movement.
- The particles will move to obtain the minimum total volume and maximum contact and compact condition during any movement without any internal friction (that is the particles are under absolute lubricity).
- There will be no reaction between the particles, no gas or volatile matter evolution, and no interparticle attractive or repulsive forces present.
- No particle will change its volume or shape during movement or after compaction.

2. Furnas Model

Working on the particle size distribution model to obtain high compaction with flowability for the mortar and concrete industries, Furnas proposed a mathematical relationship for two different types of grading, over the period from 1920 to 1930 [2]. First, one with two / three / four different size graded particles, which is a discrete packing model and the second which is based on continuous particle grading without any gap in particle sizes. Mac-Zura et al. reported [3] that the discrete packing model gives better density values but with very limited workability and flow properties, whereas the continuous particle packing model resulted in improved workability at low moisture content with significant strength and compactness [1]. However, the Furnas model for continuous distribution of particles is only an extension of his model for discrete parti-

cle distribution to a multicomponent system. According to Furnas, the best packing occurs when finer particles exactly fill the voids within the larger particles. Also the CPFT (cumulative percent finer than) curve for both cases are similar; continuous distribution consists of particles representing all sizes in the size range while discrete distribution contains monodispersions of particles representing only a fraction of the sizes in the size range.

3. Andreason Model

Andreason also worked in parallel on the same problem but with a different approach. His work was based on some empirical work, and that is why his work was not accepted as a theory. However, he proposed a simple model to calculate the continuous particle size distribution.

Basically, Andreason's model for continuous particle size distribution is based on a similarity condition. For this similarity condition, Andreason defined a granulation image of the particles and described that the granulation images surrounding particles of different sizes in the distribution be similar. Therefore, the particle array surrounding each and every particle present in the distribution, regardless of its size, is exactly similar. This arrangement of particles is the similarity condition for packing as per Andreason. Now this similarity condition is a fundamental requirement in the formation of fractals. Fractal is a mathematical set which has a fractal dimension that usually exceeds its topological dimensions. Fractals are typically self-similar patterns, where self-similar means they are "the same from near as from far." Increasing the magnification (zooming in) of a fractal picture will result in exactly similar structures at the new, higher magnification as that at the lower magnification [4–5]. Continue zooming in and you see the same structures. But fractal geometry and fractal systems were not popular or accepted till the 1960s. In a continuous particle size distribution, zooming in to any magnification will result in exactly similar particle size surroundings and pack arrangements as that at any lower magnification. As per Andreason, for continuous particle size distribution, particle size variations and packing arrangements will be exactly similar at any magnification in the distribution.

Andreason assumed such a similarity condition develops in a continuous particle size distribution system and proposed [6–8] a linear equation relation CPFT (Cumulative Percent Finer Than) plot against particle size. His equation is:

$$\text{CPFT}/100 = (D/D_L)^q$$

where:

- D - particle size, in diameter
- D_L - the largest particle size
- q - the distribution coefficient

The similarity condition is a characteristic of any particle size distribution that follows this equation. On the basis of this theoretical concept and equation, working with many experimental distributions, Andreason concluded that the exponent (q) in the equation plays a vital role in defining the particles sizes. Andreason proposed the value of the distribution coefficient to be between 0.33 and 0.5 to obtain the optimum packing density.

The drawback to Andreason's approach and equation is that he did not recognize the effect of smallest particle size [9–10]. Straight lines on log-log plots continue forever. They can reach huge numbers on the one extreme, and can reach especially small values at the other extreme. For

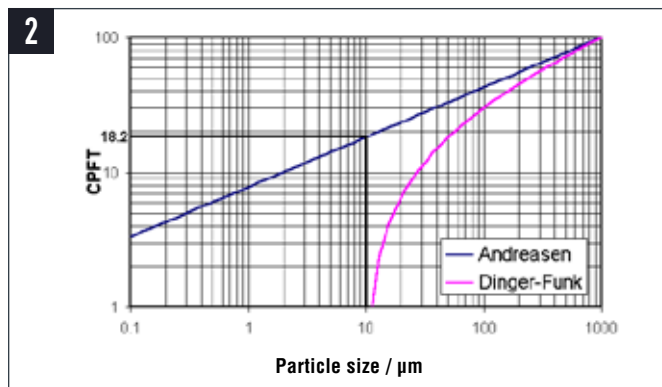


Fig. 2 • Andreason and Dinger-Funk Particle Size Distributions with $q = 0.37$, $D_L = 1000 \mu\text{m}$ and $D_S = 10 \mu\text{m}$ [15]

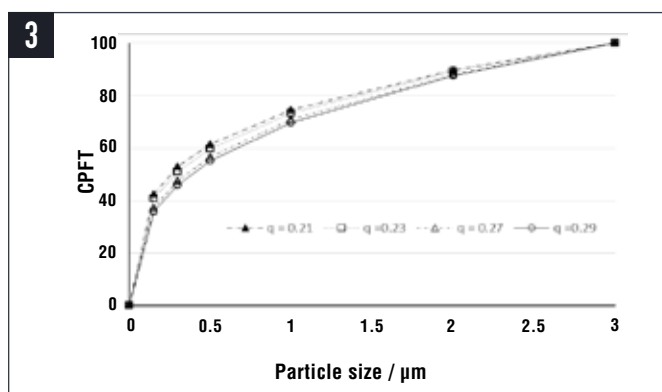


Fig. 3 • Variation of cumulative percent finer than (CPFT) with distribution coefficient (q values), having a largest size of 3 mm and finest size of 0.001 mm

example, Andreason's equation allows calculation of the CPFT of a particle having a sub-nm diameter. There will be a certain CPFT value for particles having such minute dimensions. But getting particles of that infinitesimal range and maintaining some distribution pattern is practically impossible. Hence, practically attaining any small size is not feasible and this drawback precludes Andreason's model from wide popularity.

4. Dinger and Funk Model

Experimenting with the packing of particles using Andreason's equation one can fill a certain fixed volume with particles of different size fractions going from the coarse particle to finer. To get a fully packed condition, if the volume of the space to be filled is 100 %, the volume of spheres to be packed is also 100 %. So to obtain such filling, one can simply count the number of spheres required for each different particle size used in the Andreason distribution with a particular distribution coefficient. To fill a fixed volume using this packing pattern, first the coarsest particles are packed and their number is counted. Next, the second coarsest particles are packed and their number is counted. In this way, the packing of the particles continues and the particle sizes move down through the size classes towards the finest one. Now for any practical work, the finest size (minimum available and feasible size) must be fixed and one cannot go beyond that size. So, packing is stopped at some finite minimum size, D_S , which means no finer sized particles are available to pack. Hence, for each practical system, this D_S is defined. And Dinger and Funk worked in exactly this way and defined the D_S for

the calculation of particle size distribution, which is known as the modified Andreasen equation, or Dinger and Funk particle size distribution equation [11–14]. But it took about 60 years from Andreasen's work for this modification to make the particle size distribution practically feasible. Dinger and Funk modified Andreasen's equation as follows:

$$\text{CPFT}/100 = (D^n - D_5^n) / (D_L^n - D_5^n)$$

Particle size histograms as per the Dinger and Funk equation are effectively identical to Andreasen histograms but with slightly higher percentage values (of each particle size class) due to normalization to 100 % total volume after the smallest size, D_5 , is defined. When Andreasen distribution and Dinger and Funk distribution are studied in parallel with the same distribution coefficient, q , the plots will show higher CPFT values for any size in the Andreasen equation compared to that of Dinger and Funk. Hence, Andreasen equation shows a finer distribution. Figure 2 shows such a comparison between these two models with $q = 0.37$, $D_L = 1000 \mu\text{m}$, and $D_5 = 10 \mu\text{m}$. The figure shows that there are no particles present below the 10 micron size for Dinger and Funk's model whereas 18.2 % particles are present in Andreasen's model.

The most important aspect in the packing models is that they are based on volume of the particles (involving diameters in the equations). So, whenever two or more components are present (having different specific gravity values) whilst making a castable composition, the amount of components must be converted into their volume percentages to suit the model accurately. Also, these models have been developed assuming the particles as incompressible and uncrushable spheres, but for practical situations deviations are common due to the nonsphericity (angularity) of the particles, resulting in alterations from the expected rheological behavior. Also, the models give better results when a very large number of particle size fractions is used. But in reality, say for a castable manufacturer, use of a very large number of particle size fractions causes several complications related to logistic, storage, weighing, mixing, and other associated issues. By computer simulations, it was also proposed by Dinger and Funk that for a q value of 0.37 or lower, 100 % packing efficiency can be obtained for compositions with an infinite number of fractions. This is similar to the proposal of Andreasen where he had proposed that such a packing occurs when the q value is between $1/3$ and $1/2$.

Further work on the particle size distribution models has shown that for good flowability of the castables the distribution coefficient (q) must be between 0.2 to 0.3 [16]. Also studies showed that q values close to 0.3 result in vibratable castables (requiring external energy to flow well) and lower q values (<0.25) result in castables with self-flowability (that move under their own weight) [9]. It has also been observed that higher q values result in higher coarser fractions and lower q values result in a greater finer fraction [17–20]. Figure 3 describes such a situation, decreasing q values increases the CPFT value for the sizes, thus increases the fineness of the castable granular composition. Higher fineness increases the sphericity of the particles, resulting in better flowability of the compositions and finally producing self-flowability in it. Thus by changing the q values, one can change the particle size distribution of the castable composition and control the flow behavior and other properties.

5. Conclusions

Any incorrect particle size distribution in castables will affect flow behavior and may cause dilatancy or require higher water for placement. Distribution and the amount of fine fractions present are very important

as they decide the flow and rheological behavior of the castable. The distribution coefficient (q value) plays a vital role in designing the flow behavior of the castable and it controls the amount of coarse or fine fraction to be used. By controlling the q value of the distribution, using the same particle sizes in the castable, one can obtain either a self-flowing or vibratable nature in the castable. The continuous particle size distribution model, as proposed by Dinger and Funk which considers the minimum particle size, is of practical importance and can be used for the development of castables with different flow consistencies.

References

- [1] **Homeny, J., Bradt, R.C.:** Aggregate distribution effects on the mechanical properties and thermal shock behavior of model monolithic refractory systems. In: Fisher R, ed. *Advances in Ceramics* Vol. 13. Columbus, OH: The American Ceramic Society (1985) 110–130
- [2] **Furnas, C.C.:** Grading Aggregates. *Industrial and Engineering Chemistry* **23** (1931) [7] 1052–1058
- [3] **MacZura, G., Gnauck, V., Rothenbuehler, P.:** In: Preprint of the First International Conference on Refractories. The Tech. Asso. of Refrac. (1983) 560–575
- [4] **Mandelbrot, B.B.:** A fractal set is one for which the fractal dimension strictly exceeds the topological dimension. In *Fractals and Chaos*. Springer, Berlin (2004) 38
- [5] **Gouyet, J.F.:** *Physics and fractal structures*. Masson Springer. Paris/New York (1996)
- [6] **Andreasen, A.H.M.:** Zur Kenntnis des Mahlgutes. *Kolloidchemische Beihefte* **27** (1928) [6–12] 349–458
- [7] **Andreasen, A.H.M.:** Über die Gültigkeit des Stokes'schen Gesetzes für nicht kugelförmige Teilchen. *Kolloid-Zeitschrift* **48** (1929) [2] 175–179
- [8] **Andreasen, A.H.M.:** Ueber die Beziehung zwischen Kornabstufung und Zwischenraum in Produkten aus losen Körnern (mit einigen Experimenten). *Kolloid-Zeitschrift* **50** (1930) [3] 217–228
- [9] **Myhre, B., Hundere, A.M.:** The use of particle size distribution in the development of refractory castable, XXV ALAFAR Congress, Argentina (1996)
- [10] **Dinger, R., Funk, J.E.:** Particle packing II – Review of packing of polydisperse particle systems. *Interceram* **41** (1982) [2] 95–97
- [11] **Dinger, R., Funk, J.E.:** Particle packing I – Fundamental of particle packing monodisperse spheres. *Interceram* **41** (1992) [2] 10–14
- [12] **Dinger, R., Funk, J.E.:** Particle packing II – Review of packing of polydisperse particle systems. *Interceram* **41** (1992) [2] 95–97
- [13] **Dinger, D.R., Funk, J.E.:** Particle packing III – Discrete vs continuous particle sizes. *Interceram* **41** (1992) [5] 332–335
- [14] **Dinger, D.R., Funk, J.E.:** Particle packing IV – Computer modeling of particle packing phenomena. *Interceram* **42** (1993) [3] 150–152
- [15] **Dinger, D.R.:** *Ceramic Processing E-Zine*. **1** (2003) [9] (<http://www.dingerceramics.com/CeramicProcessingE-zine/CPEBackIssues/Vol1Num9.htm>)
- [16] **Myhre, B., Sunde, K.:** Alumina based castables with very low content of hydraulic compound, Part 1: The effect of binder and PSD on flow and set. UNITECR 1995, Kyoto, Japan
- [17] **Sarkar, R., Parija, A.:** Effect of alumina fines on vibratable high alumina low cement castable. *Interceram* **63** (2014) [3] 113–116
- [18] **Sarkar, R., Parija, A.:** Effect of Alumina Fines on High Alumina Self-flow Low Cement Castables. *Refractories World Forum* **6** (2014) [1] 73–77
- [19] **Sarkar, R., Parija, A.:** Low cement high alumina castable: Effect of distribution co-efficient. International Conference on Advances in Refractories and Clean Steel Making, June 26–28, 2013, RDCIS, Ranchi, India
- [20] **Sarkar, R.:** Particle size distribution and distribution coefficient of castables. Invited talk, National Seminar on Advances in Refractory Raw Materials and Monolithics (ARMM 2013), November 12–13, (2013), Kolkata, India