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# Optimal size distribution to obtain the densest packing: A different approach

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**Abstract**

Historically a number of models were proposed about the problem of the best size distribution for densest particle packing. In general, they show a weak point in the insufficient consideration of particle morphology that, on the contrary, it is a fundamental parameter to shape an optimal size distribution. In the present work a different approach with respect the traditional models is proposed. By means of a mixture design it was possible to define models able to predict the density in function of size distribution. The results shown the effect of particles morphology: mixtures rich in fine particles permit to obtain the densest packing. The mathematical models obtained, in the form of polynomial, fit well the experimental data.


*Keywords:* Packing; Porosity; DOE

## 1. Introduction

The bulk density is strongly depending on the way particles are packed therefore there is no unique value for a given powder.[1] Generally, there are two types of packing, ordered and random. The first is packing where the particles occupy specific sites with respect to each other such that long-range order exists. The latter can be further subdivided into two types: random dense and random loose packing. In random dense packing the particles have been agitated to attain the closest packing possible without long-range order and it is equivalent to the tapped bulk density. Random loose packing is the lowest stable packing density without introducing long-range order or deformation, and it is equivalent to the aerated bulk density.[1] To obtain dense random packing is an argument of both theoretical and practical interest in a vast number of technical disciplines. Since in many ceramic products, a high sintered density and uniform grain size are highly desirable,[2] the packing efficiency is important because of its direct effect on the density of the body after the firing process. In the past, much work has gone into the studies of particle size distribution for densest packing, pore size distribution and permeability.[3] Two are the authors that have developed basic studies about this topic: Andreasen and Andersen[4] and Furnas.[5,6] Furnas' treatment is a discrete approach: best packing occurs when finer particles exactly fill the void space within the larges particles.[3] Andreasen based his model on a continuous distribution and that infinitely small particles are required to achieve the theoretically densest packing.[7] The limit of the Andreasen's model is that, in real size distribution, a minimum particle size is always present, so Funk and Dinger modified Andreasen's model introducing this information in the Funk–Dinger equation. In spite of the work that a lot of researchers have done on the argument, conflicting results from many laboratories and manufacturing experiences have led to some scepticisms about the efficiency of such methods.[3] In a previous work, the author has shown that the efficiency of classical models is quite good when the morphology of particles is enough close to a sphere, but they fail when the particles have irregular shape.[8] To overcome this obstacle, a mixture experimental design[9] with three replications has been carried out to determine the optimal size distribution for densest packing. Already Standish and Yu have shown the efficiency of this approach to study random loose packing.[10,11] The considered systems, milled glass; granulated and spray-dried aggregates, have different morphology and densities. The first sample was chosen for the irregular shape of its particles; latter two have aggregates with higher sphericity grade. The work is divided in three parts. In the first one the morphology of particles is considered. In the following section, the experimental results are discussed and mathematical models are defined using

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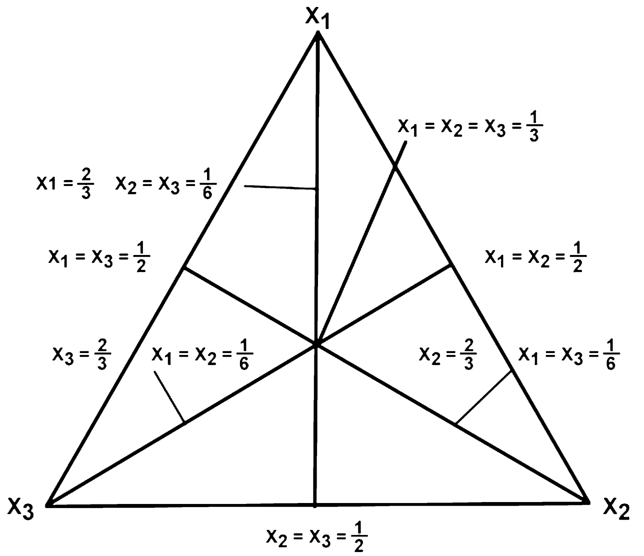


Fig. 1. Composition of the mixtures: fine ($X_1$), medium ($X_2$) and coarse ($X_3$).

mixture design. In the last part, the models are experimentally verified.

## 2. Experimental

The samples used were: milled glass; industrial spray-dried and granulated powders. For each material, three different size distributions were obtained by dry sieving: fine <250 μm; 250 μm < medium < 1000 μm; 1000 μm < coarse < 1250 μm. The software UTHSCSA ImageTool ver.3.00 was used to determine the roundness[12] of the aggregates according with Eq. (1)

$$R = \frac{4\pi A}{P^2} \tag{1}$$

where $A$ is the area of the particle and $P$ is the perimeter of the particle.

Mixtures with different percentages of fine, medium and coarse were prepared according to the scheme of Fig. 1 and their tap densities were determined (ASTM standard D4164). Three measurements for each sample were carried out. The results were analysed by the software Design-Expert 6.0.10 (Stat-Easy INC.).

## 3. Results and discussion

### 3.1. The roundness of the particles

Spray-dried and granulates aggregates are obtained with different techniques so the roundness of the particles reflect the different genesis, as it is shown in Table 1. The spray-dried powders show the highest sphericity while the glass particles have the most irregular shape. Differences are also present inside each material: in general medium sizes present the highest roundness in comparison with coarse and fine.

Table 1
Roundness for spray-dried, granulated and glass particles

| Material | Size | Average roundness |
|---|---|---|
| Spray-dried | Coarse | 0.66 |
| | Medium | 0.80 |
| | Fine | 0.82 |
| Granulated | Coarse | 0.66 |
| | Medium | 0.74 |
| | Fine | 0.65 |
| Glass | Coarse | 0.47 |
| | Medium | 0.64 |
| | Fine | 0.51 |

### 3.2. Results of mixture design and models

In Table 2 the results of the tap densities experimentally obtained are presented. Glass (GL) is the densest with respect to granulated (GR) and spray-dried (SD) particles. The explanation is that the first one is constituted of primary particles while the latter two from aggregates of primary particles.

In all the cases, quadratic is the highest order model with significant terms. By ANOVA analysis, the insignificant terms were removed. $F$-test and lack-of-fit confirmed the applicability of the model. In Table 3 statistical summary is presented.

Standard linear regression techniques (least squares) were used to fit the model to the data. The Eq. (2) represents the polynomial for granulated

$$\begin{aligned} D_{\mathrm{Gr}} = {} & 1.17 \times 10^{-2} X_1 + 1.27 \times 10^{-2} X_2 + 1.47 \times 10^{-2} X_3 \\ & + 4.09 \times 10^{-5} X_1 X_2 + 1.07 \times 10^{-4} X_1 X_3 \\ & + 6.64 \times 10^{-5} X_2 X_3 \end{aligned} \tag{2}$$

where $X_1$ is the percentage of coarse, $X_2$ is percentage of medium, and $X_3$ is the percentage of fine.

In Fig. 2 the 3D surface plot for granulated particles is presented. Tap density increases with decreasing of particle size. The lowest values are obtained when the percentage of coarse in the mixture is prevalent, while the higher tap densities are localizable in the portion of the plot richest of fine. Synergic interactions are evident among all the three size. The deviation to the linearity is stronger between fine and coarse and more limited between coarse and medium. The Eq. (3) represents the polynomial for spray-dried aggregates

$$\begin{aligned} D_{\mathrm{Atm}} = {} & 9.97 \times 10^{-3} X_1 + 1.08 \times 10^{-2} X_2 + 1.14 \times 10^{-2} X_3 \\ & + 3.75 \times 10^{-5} X_1 X_2 + 6.09 \times 10^{-5} X_1 X_3 \end{aligned} \tag{3}$$

In Fig. 3 the 3D surface plot of Eq. (3), for spray-dried particles. It shows that the tap density is influenced by the percentage of the three sizes. It increases with decreasing of particle size, but the higher densities are localizable on the axis coarse–fine. Synergic interactions are also evident between coarse–medium mixtures. The Eq. (4) is the mathematical model for glass pow-

Table 2
Compositions and tap densities for granulated, spray-dried and glass particles

| Coarse (wt.%) | Medium (wt.%) | Fine (wt.%) | Density SD (g/ml) | Density GR (g/ml) | Density GL (g/ml) |
|---|---|---|---|---|---|
| 50.00 | 0.00 | 50.00 | 1.15 | 1.58 | 1.85 |
| 0.00 | 0.00 | 100.00 | 1.11 | 1.49 | 1.52 |
| 0.00 | 100.00 | 0.00 | 1.08 | 1.26 | 1.42 |
| 100.00 | 0.00 | 0.00 | 0.99 | 1.17 | 1.41 |
| 0.00 | 100.00 | 0.00 | 1.07 | 1.28 | 1.40 |
| 16.67 | 16.67 | 66.67 | 1.21 | 1.56 | 1.72 |
| 50.00 | 50.00 | 0.00 | 1.11 | 1.31 | 1.49 |
| 16.67 | 66.67 | 16.67 | 1.10 | 1.44 | 1.61 |
| 100.00 | 0.00 | 0.00 | 1.00 | 1.17 | 1.40 |
| 0.00 | 50.00 | 50.00 | 1.16 | 1.54 | 1.77 |
| 66.67 | 16.67 | 16.67 | 1.18 | 1.46 | 1.67 |
| 33.33 | 33.33 | 33.33 | 1.23 | 1.54 | 1.81 |
| 50.00 | 0.00 | 50.00 | 1.25 | 1.59 | 1.85 |
| 0.00 | 0.00 | 100.00 | 1.14 | 1.45 | 1.52 |

Table 3
Model summary statistics for granulated, spray-dried and glass particles

| Material | Source | S.D. | Adjusted $R^2$ | Predicted $R^2$ | $R^2$ | Press |
|---|---|---|---|---|---|---|
| Granulated | Quadratic | 0.021 | 0.9796 | 0.9618 | 0.9874 | 0.011 |
| Spray-dried | Quadratic | 0.041 | 0.7182 | 0.5303 | 0.8049 | 0.037 |
| Glass | Quadratic | 0.011 | 0.9773 | 0.9617 | 0.9860 | 0.0028 |

der and Fig. 4 the 3D surface plot

$$D_{\mathrm{GL}} = \frac{1}{\sqrt[1.22]{6.61\times10^{-3}X_1 + 6.58\times10^{-3}X_2 + 6.03\times10^{-3}X_3 - 1.92\times10^{-5}X_1X_2 - 6.36\times10^{-5}X_1X_3 - 5.05\times10^{-5}X_2X_3}} \quad (4)$$

Fine particles permit to achieve the highest density while medium and coarse have similar values. Strong synergies exist between medium and fine, as well as fine and coarse, while mixtures between medium and coarse do not show remarkable increases of density. The highest values of densities are obtained in the samples where fine and coarse percentages are, respectively, about 50% and 40%. This result is shared also for spray-dried and granulated. With the increase of roundness, the importance of medium size decreases until to be unnecessary for the spray-dried.

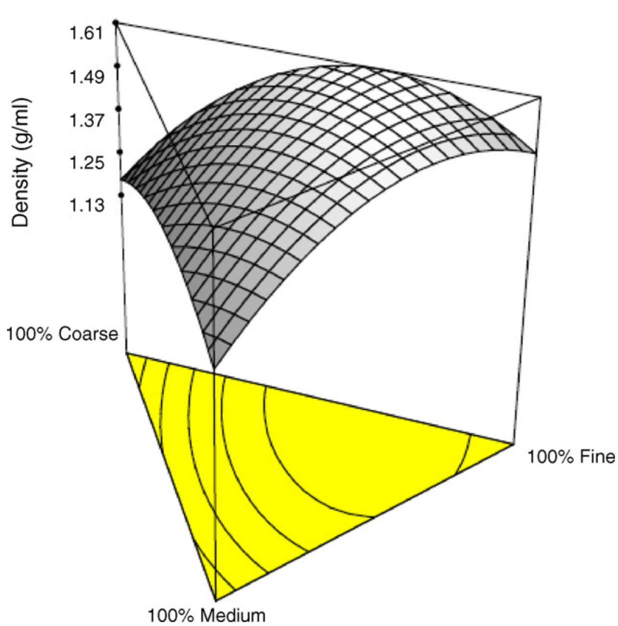


Fig. 2. 3D surface plot for granulated powder.

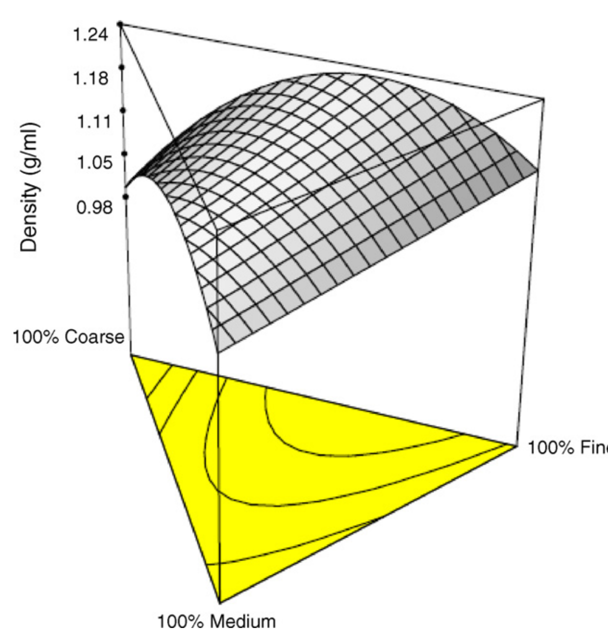


Fig. 3. 3D surface plot for spray-dried powder.

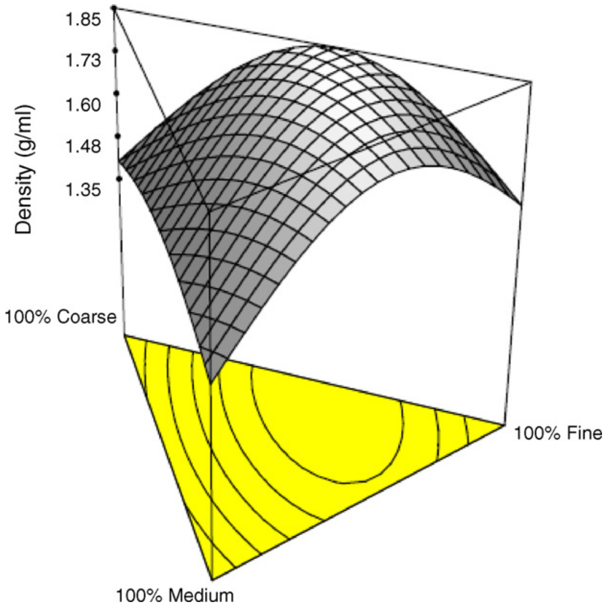


Fig. 4. 3D surface plot for glass powder.

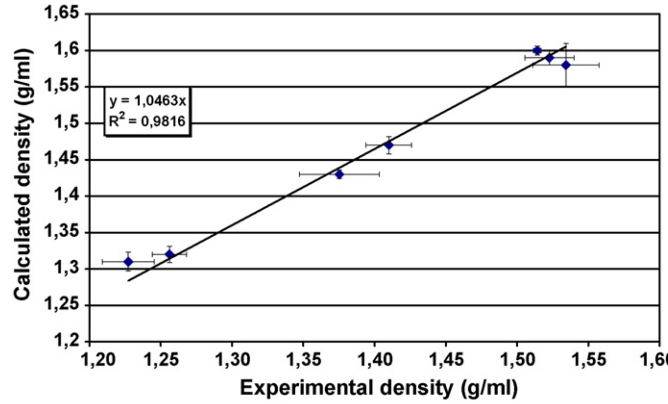


Fig. 5. Correlation between experimental and calculated (Eq. (2)) densities for granulated powders.

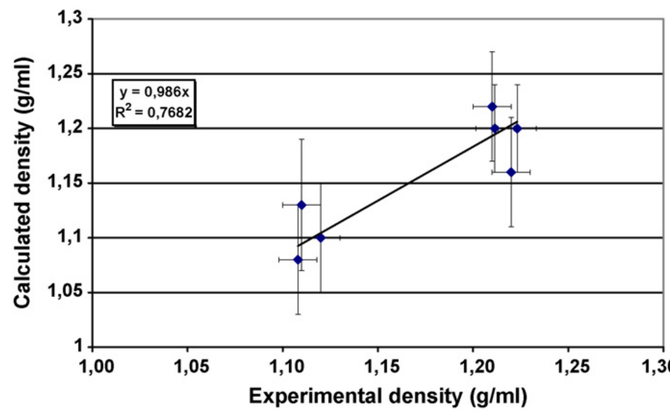


Fig. 6. Correlation between experimental and calculated (Eq. (3)) densities for spray-dried powders.

The results obtained for binary mixtures do not diverge by the results shown in the work of Westman and Hugill.[13] They asserted, and this work confirms that result, that the apparent volume is determined by the prevailing fraction present. The apparent volume reaches to a minimum, and consequently the density to a maximum, when a minority percentage of finer is present. On the contrary, in this work, the experimental results shown that the maximum is obtained when the fine size is in highest percentage. Mixtures of coarse and medium are less efficient due to the nearness between the two sizes and the morphology of the particles. For this reason a very limited synergy is observable.

### 3.3. Models check

With the purpose to check the models, seven mixtures with a composition randomly determined were prepared and the tap densities were measured as seen for previous samples, see Table 4.

The correlation between experimental and calculated values for granulated particles is plotted in Fig. 5. The slope is very close to one and the value of the coefficient of correlation $R^2$, equal to 0.9816, is high. From a statistical point of view, the probability that an equal number of random numbers have the same coefficient of correlation is less than 0.6%. For spray-dried, in Fig. 6, the slope is still close to one and $R^2$ is a bit worse. However, the probability that a equal number of random numbers have the same coefficient of correlation increases slightly up to 4.4%. For glass the coefficient of correlation goes up again to 0.9 (Fig. 7). In all the cases, the correlation is statistically significant; in particular for granulated and glass it is highly significant.

These results prove that it is possible, using DOE approach, to define models that permit not only to study the significant parameters and their interactions, or the size distribution for the densest packing, but also to predict the density of samples with a

Table 4
The compositions, experimental tap densities and calculated tap densities according to the Eq. (2) for granulated, Eq. (3) for spray-dried and Eq. (4) for glass particles

| Coarse (wt.%) | Medium (wt.%) | Fine (wt.%) | Tap density$_{exp.}$ GR (g/ml) | Tap density$_{cal.}$ GR (g/ml) | Tap density$_{exp.}$ SD (g/ml) | Tap density$_{cal.}$ SD (g/ml) | Tap density$_{exp.}$ GL (g/ml) | Tap density$_{cal.}$ GL (g/ml) |
|---|---|---|---|---|---|---|---|---|
| 32 | 21 | 47 | 1.53 ± 0.02 | 1.58 ± 0.03 | 1.22 ± 0.01 | 1.20 ± 0.04 | 1.83 ± 0.01 | 1.84 ± 0.04 |
| 26 | 20 | 54 | 1.52 ± 0.02 | 1.59 ± 0.03 | 1.21 ± 0.01 | 1.20 ± 0.04 | 1.79 ± 0.01 | 1.84 ± 0.04 |
| 83 | 9 | 8 | 1.23 ± 0.02 | 1.31 ± 0.03 | 1.11 ± 0.01 | 1.08 ± 0.05 | 1.52 ± 0.01 | 1.53 ± 0.03 |
| 72 | 0 | 28 | 1.41 ± 0.02 | 1.47 ± 0.03 | 1.22 ± 0.01 | 1.16 ± 0.05 | 1.69 ± 0.01 | 1.78 ± 0.04 |
| 26 | 0 | 74 | 1.51 ± 0.002 | 1.60 ± 0.03 | 1.21 ± 0.01 | 1.22 ± 0.05 | 1.64 ± 0.01 | 1.61 ± 0.05 |
| 0 | 78 | 22 | 1.38 ± 0.03 | 1.43 ± 0.04 | 1.12 ± 0.01 | 1.10 ± 0.05 | 1.49 ± 0.01 | 1.47 ± 0.04 |
| 25 | 75 | 0 | 1.26 ± 0.01 | 1.32 ± 0.04 | 1.11 ± 0.01 | 1.13 ± 0.06 | 1.75 ± 0.01 | 1.72 ± 0.04 |

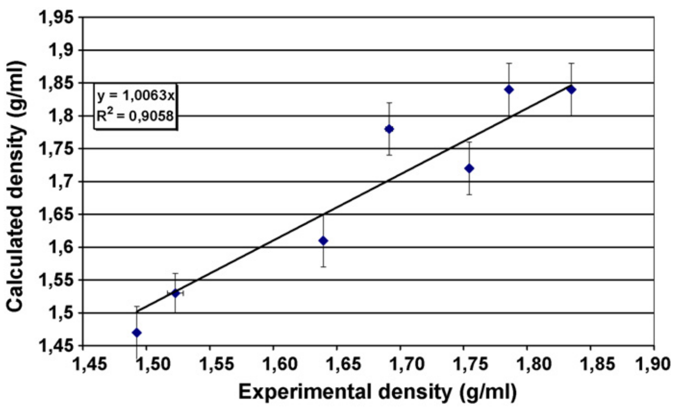


Fig. 7. Correlation between experimental and calculated (Eq. (4)) densities for glass powders.

Table 5
Calculated densest packing mixtures for granulated, spray-dried and glass particles

| | Coarse | Medium | Fine | Density (g/ml) |
|---|---|---|---|---|
| Granulated | 36.19 | 2.92 | 60.89 | 1.61 |
| Spray-dried | 38.12 | 0.00 | 61.88 | 1.23 |
| Glass | 26.95 | 0.00 | 73.05 | 1.78 |

given size distribution. On the other hands, the models are built on experimental data and the results are specific for the considered systems and not always exportable. However, the results are less subject to the approximation due to the morphology. In the present case, the best packing for glass, granulated and spray-dried is obtained, see Table 5, with quite similar compositions.

## 4. Conclusion

The tap density and mixture design were used to study the influences of size distribution on random dense packing of aggregates and primary particles, with different morphologies. Mixture design approach turned out to be a powerful tool to study the packing of non-spherical particles. According to the number of experiments carried out, the quadratic model is resulted the best model to describe the three systems. Its ability to predict the density of a sample with a known size distribution was verified to be statistically considerable. Synergies among the size classes were pointed out. The morphology of the aggregates have repercussion on the compositions with densest packing. The maximum is shifted toward high percentage of fine size. The mathematical models permit to preview the density of the respective systems, with respect to the size distribution, with a good efficiency. The limit due to the necessity to spend time at the experimental stage is paid by the efficiency of the model.

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