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Modelling of packing density for particle composites design

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Abstract

Effective packing of solid particles is one of the main topics in the field of ceramics, powder metallurgy and concrete technology. In these material sectors it is necessary to maximise or optimise the packing density of particles. Therefore, it is necessary to obtain the ability not even to measure the packing density effectively but especially to predict it and affect it with sufficient accuracy. Despite of large experiences in field of metallurgy and ceramics technology, it is still relatively difficult to predict packing density in the concrete technology. Prediction is based on de Larrard linear packing theory expanded by third parameter including wedging effect of particles to the form of 3-parameter packing model. In this paper the model is calibrated for fillers using in Particle composites technology with respect to their granulometry, mainly aimed on UHPC technology. Calibration is based on correlation with experimentally determined values of packing density of model particles mixtures. Successful optimization of particular system composition in concrete technology then could lead not even to decrease of final price but it has also a beneficial influence mainly on mechanical properties and durability of final product.

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Keywords: Packing density; particle packing model; particle interaction; mix design

1. Introduction

Particle composites such a Ultra-High Performance Concretes (UHPCs) is a group of unique inorganic composites which are characterized by very high compressive strength over 150 MPa, high durability estimated at 200 years and nearly no permeability for corrosive agents. Besides other factors, these properties are achieved by precise selection of raw materials with respect to their particle shape, particle size and granulometry to reach highly

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compact structure with minimum free volume, with maximum packing density, respectively [1]. In spite of large experiences of particle packing in the field of ceramics, powder metallurgy or pharmacy, in modern concrete technology it is still relatively difficult to predict and effectively influence the particle packing density. Particle packing has a fundamental influence on final properties of solid particular materials, such as rheology of prepared suspensions, porosity, permeability or strength. Therefore it is necessary to understand particle packing mechanisms. If we assume a particular system consisting of same size sphere particles, it is easy to calculate that the packing density of such a system is 74.05% (hexagonal close package). Unfortunately, in fact, it is not possible to reach such a high value with mentioned system where maximum packing density is not higher than 64% [2]. For increasing the packing density it is necessary to add such a particle which fill the gap between coarser particles. Fundamentally, the particle packing theory can be divided into two main branches.

First of them is continual packing theory, firstly introduced by Fuller and Thompson and modified by Dinger and Funk to modern form known as Modified Anders-Andreassen model. Continual theory considers that particles are packed continuously according to decreasing particle size [3]. From mathematical formulation of mentioned models is evident that the highest grade of packing is reached when the cumulative granulometric curve of particular system is close to be parabolic. Unfortunately, real systems are not able to pack in such a way in real time due to irregularities in packing. Also real systems has not continuous grading, because they are consist of particles with discrete size. It is only possible to get close to this ideal grading which is mainly limited by number of raw materials stored in real production facility.

Limitations of continual packing theory can be partially solved by using of discrete packing theory. In this theory the particular system is divided to the discrete particle size classes where at least one class is dominant and it is packed preferably. The others particle size classes are then packed to this skeleton. First discrete packing model was introduced by Furnas and Westman on binary systems where particle size ratio was near to zero. In this model there were defied two effect which increase packing density; namely filling effect of fine particles in gaps between coarse particle class and occupying effect of coarse particles which replace the porous space of fine particle size [1,4].

When the particle size ratio is not in limit case (0 or 1), irregularities which decrease packing density take place. These irregularities, also called packing restrictions were firstly introduced by Stovall in the form of two structural effects which disrupt the regular packing and therefore decrease the packing density; namely loosening effect of the fine particle class which disrupt the packing of dominant coarse particle class by squeezing themselves between them; and wall effect of the coarse particle class by disrupting of dominant fine particle class packing by forming wall-like structures on boundaries between particle classes. Participation rate of each effect depends on diameter ratio of interacted particle class and it is presented as interaction function of diameter ratio of two particle classes where one is dominant class. Several authors (de Larrard, Yu) derived their own interaction function and incorporated them to the form of so called Linear Packing Density Model [3,4].

Dependence of packing density on volume percentage of fine particle class is actually not linear, as many authors suggest (de Larrard, Kwan) [4,5]. Especially in the optimum composition point where linear packing theory overestimate the packing density i. e. there should be another effect which decrease the packing density especially around the optimum. This effect was called wedging effect and it was firstly introduced by Kwan. Wedging effect occurs in both cases, when the dominant particle class interact with fine particle class and also with coarse particle class. In the case where the dominant particle class interact with finer particles where the fine particles can be wedged between the coarser particles instead of to fill the space between them. It results in displacement of coarse particles and therefore decrease of packing density. Contribution of wedging effect is higher when the size of small particles is close to size of the gaps between coarse particles because there is the highest probability that particle can be wedged. Therefore, wedging effect acts at most in the optimum percentage of fine particle class. Wedging effect also occurs when the dominant class interacts with coarser particle class. These coarser particles are dispergated each other between the fine particles of dominant class but the dispergation is not homogenous. In the case when the gap between coarser particles is too small for fine particle from dominant class the incomplete layer of this particles is formed around the coarser particles which caused decrease of packing density. In other words when the fine particles from dominant class closer in size to gaps between coarser particles, the probability of incomplete layer increases. Therefore, contribution of wedging effect also increases around the optimum combination [5].

2. Experimental program

This model, also known as 3-parameter model, was originally developed for binary mixture of sphere particles where only two particle size class are presented. This model can be easily described by two equation (Eq. 1a and 1b), where ϕ_1^* and ϕ_2^* are packing densities when class 1 or 2 is dominant, ϕ_1 and ϕ_2 are the packing densities of individual class 1 and 2, y_1 and y_2 are the volume fraction of particle size class 1 and 2, y_1^* and y_2^* are the optimum volumetric fraction of particle size class 1 and 2 and a_{12}, b_{12} and c_{12} are the interaction functions characterizing loosening effect, wall effect and wedging effect. Each for case where finer class or coarser class is dominant. The real packing density is than define as $\phi = \min\{\phi_1^*, \phi_2^*\}$ [6].

$$\frac{1}{\phi_1^*} = \left(\frac{y_1}{\phi_1} + \frac{y_2}{\phi_2}\right) - (1-b_{12})(1-\phi_2)\frac{y_2}{\phi_2}\left[1 - c_{12}\left(\frac{y_2}{y_2^*}\right)\right] \tag{1a}$$

$$\frac{1}{\phi_2^*} = \left(\frac{y_1}{\phi_1} + \frac{y_2}{\phi_2}\right) - (1-a_{12})\frac{y_1}{\phi_1}\left[1 - c_{12}\left(\frac{y_1}{y_1^*}\right)\right] \tag{1b}$$

Interaction function were experimentally determined by several authors. In this study we used those derived by Kwan, mainly due to incorporation of wedging effect. Interaction functions are described by following equation (2a, 2b, 2c)

$$a = 1 - (1-s)^{3.3} - 2.6 \cdot s \cdot (1-s)^{3.6} \tag{2a}$$

$$b = 1 - (1-s)^{1.9} - 2 \cdot s \cdot (1-s)^{6} \tag{2b}$$

$$c = 0.322 \cdot \tanh(11.9 \cdot s) \tag{2c}$$

where s is the diameter ratio in order to having a value in interval (0,1) [5,7].

As can be seen the packing density is in this form dependent on optimum volumetric fraction of class 1 and 2, respectively, which is actually parameter which we want to determined (based on maximum packing density). Therefore, Kwan modified the original 3-parameter model by substitution for y_1^* and y_2^* derived from linear packing density model. Due to this modification the packing density is no longer dependent on optimum volumetric fraction. Using this substitution Kwan has made extension of this function for ternary particle size system (system consisted of 3 particle size classes) which is represented by following three equations [5,7]:

$$\frac{1}{\phi_1^*} = \left(\frac{y_1}{\phi_1} + \frac{y_2}{\phi_2} + \frac{y_3}{\phi_3}\right) - (1-b_{12})(1-\phi_2)\frac{y_2}{\phi_2}\left[1 - c_{12}\left(2.6^{(y_2+y_3)} - 1\right)\right] \tag{3a}$$

$$\frac{1}{\phi_2^*} = \left(\frac{y_1}{\phi_1} + \frac{y_2}{\phi_2} + \frac{y_3}{\phi_3}\right) - (1-a_{12})\frac{y_1}{\phi_1}\left[1 - c_{12}\left(3.8^{y_1} - 1\right)\right] - (1-b_{23})(1-\phi_3)\frac{y_3}{\phi_3}\left[1 - c_{23}\left(2.6^{y_3} - 1\right)\right] \tag{3b}$$

$$\frac{1}{\phi_3^*} = \left(\frac{y_1}{\phi_1} + \frac{y_2}{\phi_2} + \frac{y_3}{\phi_3}\right) - (1-a_{13})\frac{y_1}{\phi_1}\left[1 - c_{13}\left(3.8^{y_1} - 1\right)\right] - (1-a_{23})\frac{y_2}{\phi_2}\left[1 - c_{23}\left(3.8^{y_2} - 1\right)\right] \tag{3c}$$

These three equations can be now directly easily used to derive a generalized form for multimodal particular system consisted of n particle size classes. If we assume that each individual particle class consists of particles with the same size, we can transfer values of $\phi_1, \phi_2, ...\phi_n$ to one general value ϕ_C which is same for all classes in the particular system. The generalized form of 3-parameter Packing Density Model is described by Eq. 4.

$$\phi_i^* = \frac{1}{\sum_{i=1}^{n} \frac{y_i}{\phi_C} - \sum_{j=1}^{i-1} (1-a_{ij}) \frac{y_j}{\phi_C} \left[1 - c_{ij}\left(3.8^{y_j} - 1\right)\right] - \sum_{j=i+1}^{n} (1-b_{ij})(1-\phi_C) \frac{y_j}{\phi_C} \left[1 - c_{ij}\left(2.6^{\sum_{j=i+1}^{n} y_j} - 1\right)\right]} \tag{4}$$

This derived generalized packing density function based on Kwan's extension of 3-parameter Packing Density Model was primary used in this study.

The original 2-parameter packing model and extended Kwan's 3-parameter packing model were used for calculation of packing densities of sphere particles mixes with wider granulometric curve, in other words the particles mixes can by characterizes as multimodal. The results of calculation were compared to experimentally determined value of packing density of each mixes. Based on these results the calibration program was designed.

3. Materials and methods

In this study there were used four standard mixes of glass beads as sphere particles multimodal granulometric system, signed as B1, B2, B3 and B4 according to increasing median of particle size. Granulometric curves of each standard mix are shown in Fig. 1.

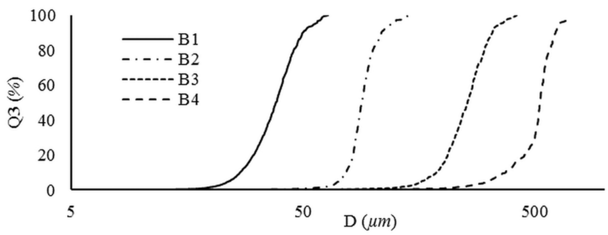

Fig. 1. Granulometry of used materials.

The granulometric curves were determined by image analysis from optical microscopy record. For each curve there were analyzed at least 1000 particles. From granulometry curves there were also determined the values of $D_{\min}$ and $D_{\max}$ as 1% quantile and 99% quantile. For each material mix there was also determined particle volume weight ρ. Material characteristic for each particle mix are summarized in Table 1. In this study there were prepared mixtures of standard glass beads mixes in combination of B1 and B3 (B1-B3), B1 and B4 (B1-B4), B2 and B3 (B2-B3) and B2 and B4 (B2-B4). For each combination there was experimentally determined the packing density in order of increasing volumetric fraction of finer constituent.

Table 1. Material parameters.

Parameter	B1	B2	B3	B4
$D_{\min}$ (μm)	19.2	56.3	124.3	216.4
$D_{\max}$ (μm)	60.9	137.4	403.5	696.2
ρ (g/cm^3)	2.473	2.475	2.476	2.475

Experimental determination of packing density was provided according to EN 1097-3:1998 with additional compaction by standard vibration table [8,9]. From measuring of compacted powder body weight M, the packing density $\phi_{\exp}$ was determined according Eq. 6,

$$\phi_{\exp} = \frac{M\left(\rho_i y_i + \rho_j y_j\right)}{V} \tag{6}$$

where V is volume of testing container, ρ_i and ρ_j is the volume weight of constituent i and j and y_i and y_j is the volumetric ration of constituent i and j. Experimentally determined packing density was plotted against the packing densities calculated using 2-parameter packing model and Kwan's extended 3-parameter packing model.

4. Results and discussion

In this study there were analyzed the packing densities of mixtures of spherical glass beads B1-B3, B1-B4, B2-B3 and B2-B4 in order of increasing volume ration of finer constituent, B1 and B2 respectively. Experimental results were plotted against calculated values according to 2-parameter packing model (linear packing density model) and Kwan's extended 3-parameter packing model. Two main criteria were observed; maximum of packing density and its position against coordinate x, optimum combination respectively. Results from first phase are graphically summarized in Fig. 2 – 5.

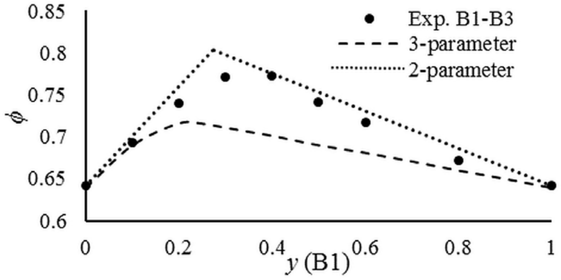

Fig. 2. Mixture B1-B3.

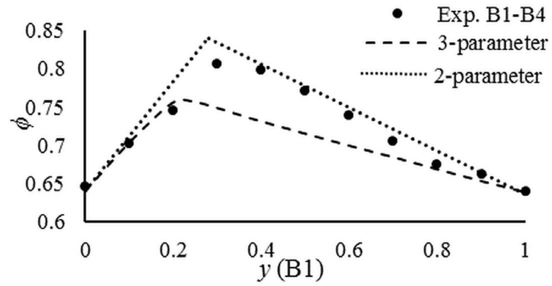

Fig. 3. Mixture B1-B4.

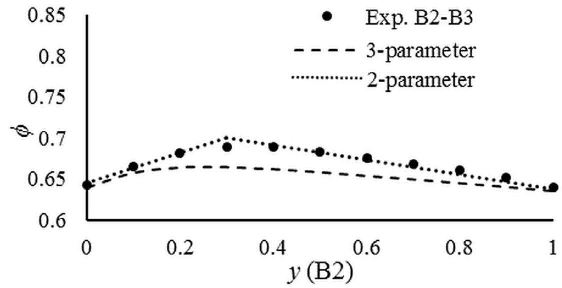

Fig. 4. Mixture B2-B3.

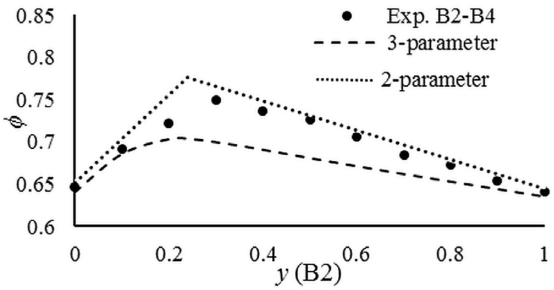

Fig. 5. Mixture B2-B4.

As can be clearly seen, prediction of packing density by Linear 2-parameter Packing model slightly overestimate the real value of packing density around the optimum combination in all tested samples. Also can be seen that the maximum position is slightly moved to lower content of finer constituent. These results fully correspond to the linear nature of 2-parameter packing models and show that the real packing density is not, especially around the optimum, linear dependence of constituent's ratio. Same conclusion, as other authors pronounced, can be expressed. There must be another effect which decrease the packing density, mainly around the optimum of constituent ratio. Using Kwan's extended 3-parameter packing model where third parameter, wedging effect, is included, can be calculated another curve. Actually, this model substantially underestimated the experimentally determined values of packing density. Moreover, the maximum position is shifted to even lower values of constituent ration than in the case of 2-parameter model. As can be seen, in ratio near to 0 or 1, where wedging effect has very low contribution, the calculated values from 3-parameter model approximately corresponds with those experimentally determined. The highest deviation can be seem round the optimum ration and higher, where the wedging effect interaction function reaches too high values, in contrast to results determined by Kwan. From our results it seems to be that more complex particle system, their multimodality respectively, has a great influence on wedging effect participation. As it was mentioned above, wedging effect occurs, when dominant particle class participate with both smaller particle classes and larger particle classes. Interaction function can be therefore solved in two separated parts. The first one when we assume participation of dominant class with smaller particle class and the second one when we assume the participation with larger particle classes, according to Eq 7 for the first case and Eq. 8 for the second case.

$$c_1 = 0.322 \cdot \tanh\left(11.9\frac{d_j}{d_i}\right) \tag{7}$$

$$c_2 = 0.322 \cdot \tanh\left(11.9\frac{d_i}{d_j}\right) \tag{8}$$

From the Eq. 4 is clear that for increasing of calculated packing density to meet the experimental values it is necessary to magnify somehow the value of interaction function for wedging effect. Therefore, it was conducted a dependence of packing density ϕ_i^* on c_1 and c_2 for constituent ratio according to experimentally determined values; namely for constituent ratio $y = \{0; 0.1; 0.2; 0.3; 0.4; 0.5; 0.6; 0.7; 0.8; 0.9; 1\}$. By this operation it was obtained a series of 11 areas. For idea on Fig. 6 – 7 are graphically summarized results for mixture B1-B3 and y = 0.4 and 0.8.

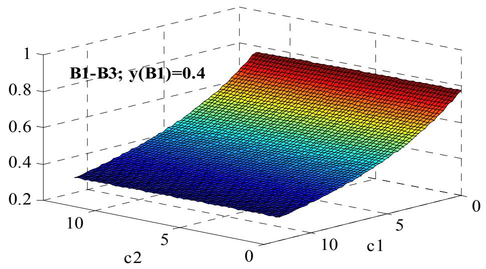

Fig. 6. B1-B3; y = 0.4.

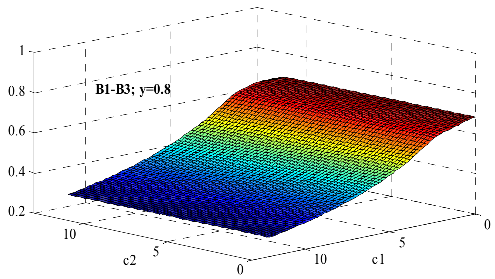

Fig. 7. B1-B3; y = 0.8.

From results can be seen that calculated packing density by 3-parameter packing model is strongly depended on parameter c_1 instead of parameter c_2 where the dependence is very low. This effect is visible in all plotted areas. To find an ideal value of c_1 and c_2 magnification coefficients k_1 and k_2 for a given mixture we have to find a series of 11 coordinates (1 for each area) corresponded to the calculated packing density which have the lowest deviation compared to relevant experimentally determined values of packing density. This can be easily done by Residual Squared Sum (*RSS*) method with 11 members generally described by Eq. 9.

$$RSS = 1 - \sum_{i=1}^{n}\left(y_i - f(x_i)\right)^2 \tag{9}$$

roviding this operation based on calculated data for mixtures B1-B3, B1-B4, B2-B3 and B2-B4 we get a series of for values of k_1 and k_2 summarized in Table 2.

Table 2. Derived magnification coefficients k_1 and k_2 for each mixture.

Parameter	B1-B3	B1-B4	B2-B3	B2-B4
k_1	9.15	10.77	0.50	5.45
k_2	0.08	0.10	0.02	0.06
RSS	0.9998	0.9995	0.9999	0.9998

There are visible a substantial difference between derived magnification values of c_1 (mainly) and c_2. The differences have to be related somehow to the shape of granulometric curve. There have to find a parameter which is same for all 9 granulometric curves belonging to complete mixture. Such a parameter could be possibly D_{min}/D_{max} ratio. Plotted D_{min}/D_{max} ratio against k_1 and k_2 respectively gives a dependence which can be fitted by linear curve according to Fig. 8 and Fig. 9. Using relevant equations to these curves the values of k_1 and k_2 can be easily calculated with relative high accuracy in investigated range of $D_{min}/D_{max.}$.

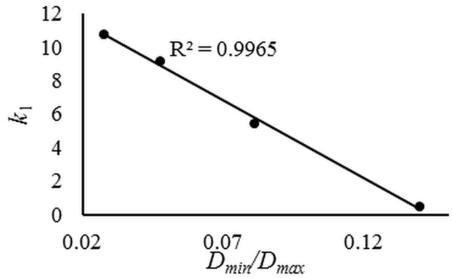

Fig. 8. Dependence of x_1 on D_{min}/D_{max}.

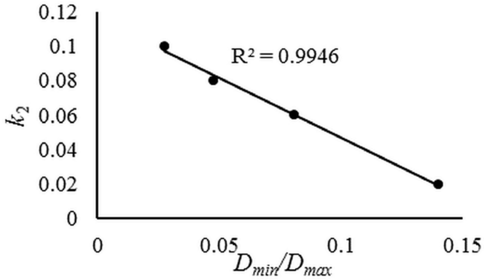

Fig. 9. Dependence of x_2 on D_{min}/D_{max}.

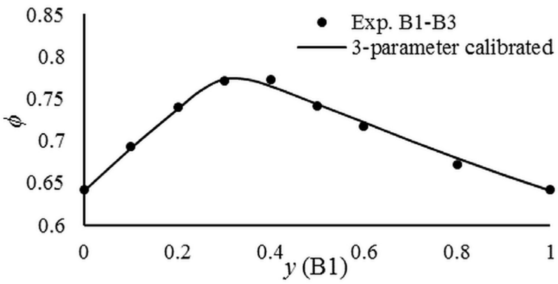

Fig. 10. B1-B3 - calibrated 3-parameter model.

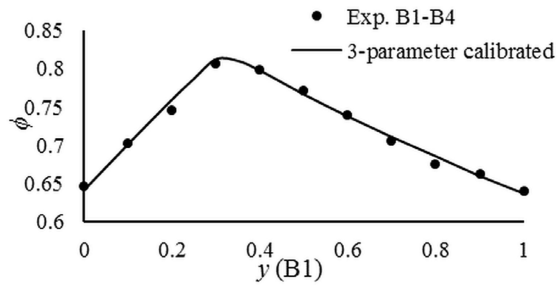

Fig. 11. B1-B4 - calibrated 3-parameter model.

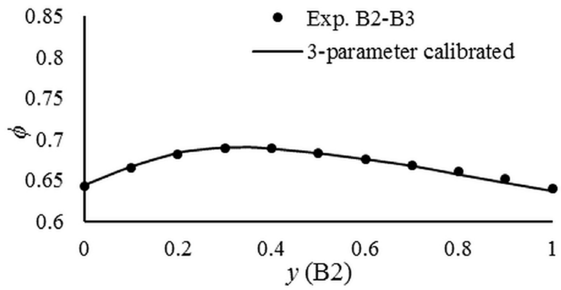

Fig. 12. B2-B3 - calibrated 3-parameter model.

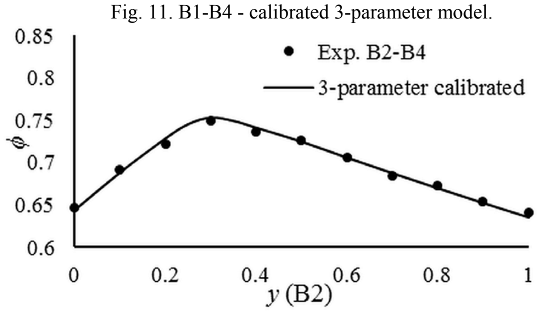

Fig. 13. B2-B4 - calibrated 3-parameter model.

Using derived magnification coefficients k_1 and k_2 in calculation algorithm of 3-parameter packing model we have got a curves according to Fig. 10 – 13. As can be seen, curves perfectly fitted the experimentally determined data within the error maximally 1% which creates a good assumption for application of this modified version of Kwan's 3-parameter model on random particular material consists of spherical particles and defined by its granulometric curve. Further investigation is needed and wider spectrum of combination and mixtures have to be tested for confirmation of our hypothesis that contribution of wedging effect is strongly depended on shape and modality of granulometric curve. Further investigation will be directed not even to confirmation of these assumption but also to influence of particle shape on packing density followed by modification of 3-parameter packing model to this factor. Successful achievement of this goal enables the direct incorporation of such a method to mix design of High Performance and Ultra-High Performance Concretes and other particle composites.

5. Conclusions

The possibilities to predict packing density of multimodal particle systems consist of spherical particles by conventional 2-parameter packing model and Kwan's extended 3-parameter packing model were investigated. Calculated results were compared to experimentally determined packing densities of binary mixtures of multimodal fraction of spherical particles. According to general assumption it was observed that prediction of packing density by conventional 2-parameter packing model is overestimated around the optimum combination. Slightly shift of optimum volume fraction was also observed. Results from original formulation of 3-parameter packing model by Kwan shows substantial deviation from experimental results. As we suppose, reason is in formulation of interaction function for wedging effect which was not originally developed for multimodal particle systems and its contribution and the value of relevant interaction function is somehow related to the shape of granulometric curve. Results from Kwan's 3-parameter model underestimate the experimental results, especially around the optimum where wedging effect has the largest contribution. Therefore, interaction function for this effect had to be magnified. We found than main contribution of wedging effect is when dominant class interact with smaller particles. By magnification of interaction function for wedging effect to approach to the experimental values of packing density, the proper magnification coefficients k_1 and k_2 was found for each mixture. Comparing magnification coefficients with D_{min}/D_{max} ration of relevant mixture we found a relationship between magnification coefficients k_1 and k_2 which has the form of linear curve. It seems to be that from this curves the value of k_1 and k_2 can be easily calculated for a particular system characterized by D_{min}/D_{max}. However, further investigation is needed, especially in the case of experimental testing of packing destiny for more granulometric systems consist of spherical particles which take place in following study. In following study there will be also investigated an influence of particle shape to packing density and the proper modification of packing density model will be proposed.

Acknowledgements

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