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O Koutny[1], J Kratochvil[1], M Drdlova[1] and E Bystrianska[1]

[1] Materials Research Centre, Brno University of Technology, Purkyňova 118,
612 00 Brno, Czech Republic

Email: xckoutnyo@fch.vut.cz

Abstract. The presented study follows the author's results of research where 3-parameter packing model for packing density determination was derived and calibrated for multimodal particular system. In this study previously derived model is extended by fourth parameter related to particle shape and its effect on bulk packing density. The model is primary derived for high performance cementitious composite design where compact structure and low binder content is preferred to maximise mechanical properties and durability. The effect of aggregate packing density on mechanical properties of ultra-high performance cementitious composites is investigated, using derived packing model

1. Introduction

Particle packing plays a key role in many fields of material engineering such as powder metallurgy, ceramics or polymer composites. Beside all of these, the cementitious composite industry utilizes the particle packing in the most common way into the production of the most world-spread construction material – concrete. The precise mixing of raw particular materials enables to achieve the structure with optimal or maximal packing density of particles. Such a dense structure improves many material properties such an increase of mechanical strength and durability, reduction of volume changes and overheating during the setting because of lower content of binder. The dense packing is the main requirement for concrete production in modern way into the form of material which is called high performance and ultra-high performance concrete (HPC and UHPC). In spite of large experiences of particle packing in the industrial fields mentioned above it is still relatively difficult to predict and effectively influence the particle packing density in modern concrete technology.

   Packing density of particular system can be analytically determined via several existing models based on 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 [1]. In this model there were defined two effects 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. 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 [2]. These effects are namely loosening effect of the fine particle class which disrupts 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) [5, 6]. 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 [7].

In the original study of authors the Kwan extended 3-parameter packing model was calibrated to predict packing density of multimodal particle systems consisting of spherical particles. Calculated results were compared to experimentally determined packing densities for binary mixtures of multimodal fraction of spherical particles. Results from original formulation of 3-parameter packing model by Kwan show substantial deviation from experimental results. The 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 have to be somehow related to the shape of granulometric curve. Therefore, interaction function for this effect has been adjusted [8].

Unfortunately, such a systems are not in coherence with real requirements of cementitious composites industry where the raw materials consist of particles with non-spherical shape. Kwan uses his original 3-parameter packing model for packing density determination of angular rock aggregate particles with correction of interaction functions for loosening effect and wall effect [9]. But there is still no direct connection between defined particle shape and change of packing. Therefore it is necessary to find out the parameter responsible for packing density change based on particle shape.

2. Experimental program
Presented study follows the results of [8], where the original Kwan extended 3-parameter packing model was calibrated to predict packing density of multimodal particle systems consisting of spherical particles of n particle size classes. The model can be described in its generalized and calibrated form by equation 1:

$$\frac{1}{\phi_i^*} = \sum_{i=1}^{n} \frac{y_i}{\phi_i} - \sum_{j=1}^{i-1} (1-a) \frac{y_j}{\phi_j} \left[1 - c\left(3.8^{y_j} - 1\right)\right] -$$
$$- \sum_{j=i+1}^{n} (1-b)(1-\phi_j) \frac{y_j}{\phi_j} \left[1 - c\left(2.6^{\sum_{j=i+1}^{n} y_j} - 1\right)\right]$$

(1)

where ϕ_C represents packing density of individual particle size class, y_i and y_i represents volume fraction of particle class size i and j respectively and parameters a, b and c represent interaction function for loosening effect, wall effect wedging effect according equation 2–5. The interaction function for wedging effect is solved separately for situation when $d_i < d_j$ using equation 4 and for situation when $d_i > d_j$ using equation 5.

$$a = 1 - \left(1 - \frac{d_i}{d_j}\right)^{3.3} - 2.6 \frac{d_i}{d_j} \left(1 - \frac{d_i}{d_j}\right)^{3.6}$$

(2)

$$b = 1 - \left(1 - \frac{d_j}{d_i}\right)^{1.9} - 2 \frac{d_j}{d_i} \left(1 - \frac{d_j}{d_i}\right)^{6}$$

(3)

$$c_1 = 0.322 \tanh\left(11.9 \frac{d_i}{d_j}\right) k_1 \qquad (4)$$

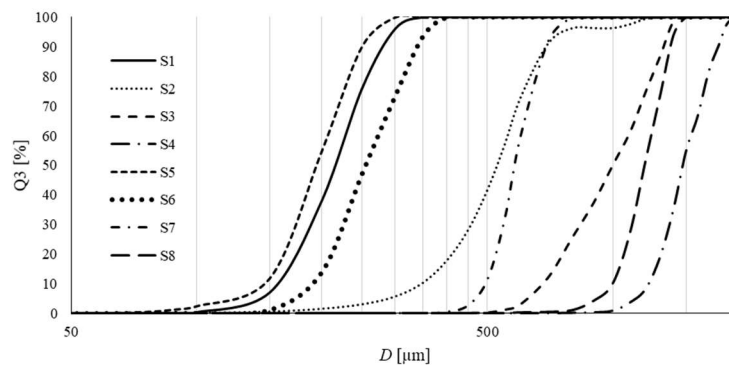$$c_2 = 0.322 \tanh\left(11.9 \frac{d_j}{d_i}\right) k_2 \qquad (5)$$

where d_i is particle diameter of size class i, d_j is particle diameter of size class j (dominant class) and k_1 and k_2 are magnification factors for wedging effect interaction function. The real packing density is then defined as $\phi = \min\{\phi_1^*, \phi_2^*, \dots\}$.

The raw materials for cementitious composites technology do not consist of spherical particles. Therefore the deviation can be expected in prediction of packing density of real particular systems using above derived model. This deviation has to be reflected and compensated by introduction of fourth parameter, as a function of packing density of individual size class ϕ_i instead of ϕ_C on parameter which characterises shape of particle in size class i.

The practical use of presented packing model for non-spherical particles is demonstrated on design of ultra-high performance cementitious composites. The effect of aggregate packing density on final mechanical properties is monitored on designed materials.

3. Materials and Methods

In this study there were used eight fractions of siliceaous aggregates as a non-spherical particle multimodal granulometric system, signed as S1, S2, S3, S4, S5, S6, S7 and S8 according to increasing median of particle size. Granulometric curves of each standard mix are shown in figure 1.

Figure 1. Granulometry of used materials.

The granulometric curves were determined using image analysis from optical microscopy record. For each curve there were analysed at least 1000 particles. The particle shape is described by circularity C of particle projection in optical microscopy record. The standardised circularity C^* is expressed as the median of C for relevant aggregate mix. From granulometric 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 characteristics for each particle mix are summarized in table 1.

Table 1. Material parameters.

	S1	**S2**	**S3**	**S4**	**S5**	**S6**	**S7**	**S8**
D_1 [µm]	133	190	548	1034	89	147	414	962
D_{50} [µm]	201	528	997	1495	198	257	579	1215
D_{99} [µm]	309	1175	1408	1944	299	413	754	1449
ρ [g/cm³]	2.67	2.67	2.65	2.64	2.66	2.64	2.63	2.64
C^*	0.834	0.876	0.884	0.883	0.816	0.842	0.846	0.855

From described materials there were prepared combined mixes of S1 and S4 (S1-S4), S1 and S3 (S1-S3), S2 and S4 (S2-S4), S2 and S3 (S2-S3), S5 and S7 (S5-S7) a S6 and S8 (S6-S8). These mixes were used for analysis and following correction of packing density. For each mix the packing density was experimentally determined in dependence on increasing volume ratio of finer standard fraction.

Experimental determination of packing density was provided according to EN 1097-3:1998 with additional compaction by standard vibration table. From measuring of compacted powder body weight M, the packing density ϕ_{exp} was determined according equation 6:

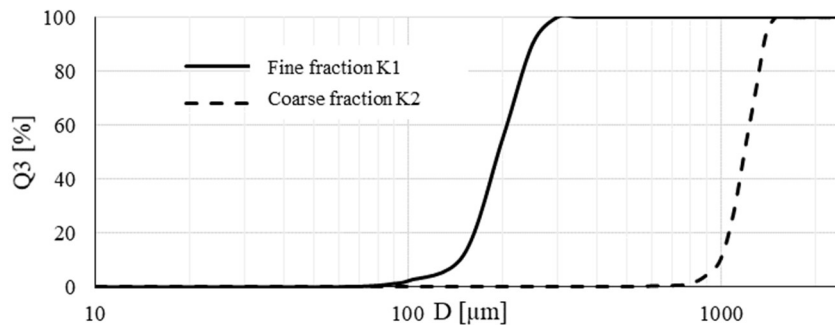$$\phi_{exp} = \frac{1}{V} M \sum_{i=1}^{n} \rho_i y_i \tag{6}$$

where V is volume of testing container, ρ_i is the volume weight of constituent i and y_i is the volumetric ration of constituent i.

Mechanical properties, the compressive strength and the flexural strength, are monitored for samples of ultra-high performance mortars which were designed with different composition of aggregate skeleton and therefore with different packing density of aggregates. Two fractions of corundum aggregates were mixed in proper volume ratios (y K1 and y K2) to reach defined packing density ϕ_{agg} according the table 2.

Table 2. Composition of control mixes.

ϕ_{agg}	0.56	0.58	0.60	0.62	0.64	0.66	0.67
y(K1)	0.021	0.073	0.124	0.179	0.233	0.297	0.369
y_{agg}	0.536	0.557	0.577	0.597	0.618	0.638	0.648
ϕ_{agg}	0.66	0.64	0.62	0.60	0.58	0.56	
y(K1)	0.426	0.528	0.619	0.720	0.822	0.923	
y_{agg}	0.638	0.618	0.597	0.577	0.557	0.536	

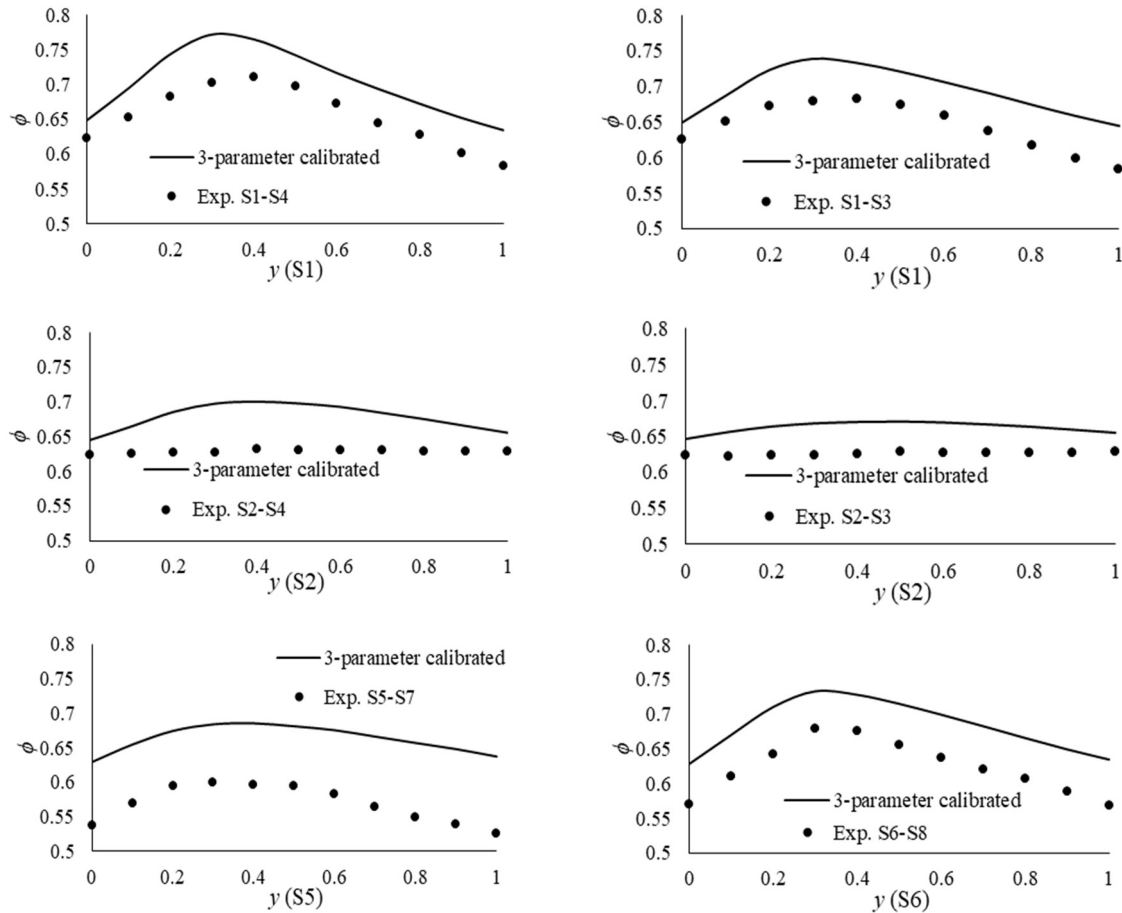The granulometry of used corundum fractions is presented in figure 2.

Figure 2. Granulometry of used corundum aggregates.

The aggregate volume fraction y_{agg} was determined as $1-0.05(1-\phi_{agg})$ where the expression $0.05(1-\phi_{agg})$ is equal to volume fraction of binder y_b. A binder based on Portland cement CEM I 52,5 R, undensified silica fume and ground limestone in mass portion of 7:2:1 was used for all mixes. The high-range water reducing agent based on polycarboxylate-ether copolymer was used for all mixes in portion 2.5% to binder. The water-to-binder ratio was set on 0.22. The test specimens were prepared in dimensions 40 x 40 x 160 mm for mechanical properties determination. Samples were compacted and cast using vibration table. The compressive strength was determined as a mean of six values and the flexural strength was determined as a mean of three values. After the mixing the workability was controlled using mini-spread flow test.
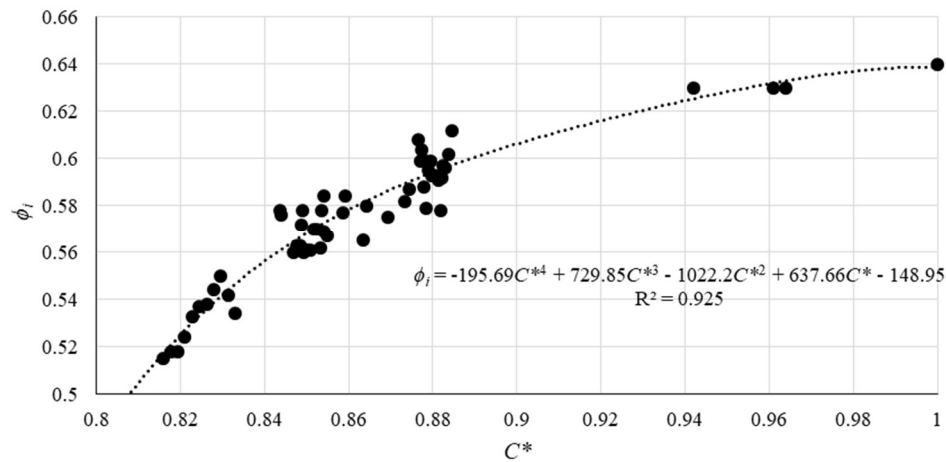
4. Results and discussion

Experimentally determined values of packing density for defined mixes were compared with values provided by calibrated modified 3-parameters model derived in previous study [8]. Two main criteria were monitored; the maximal value of packing density and corresponding composition of the mix. The results are shown in figure 3.
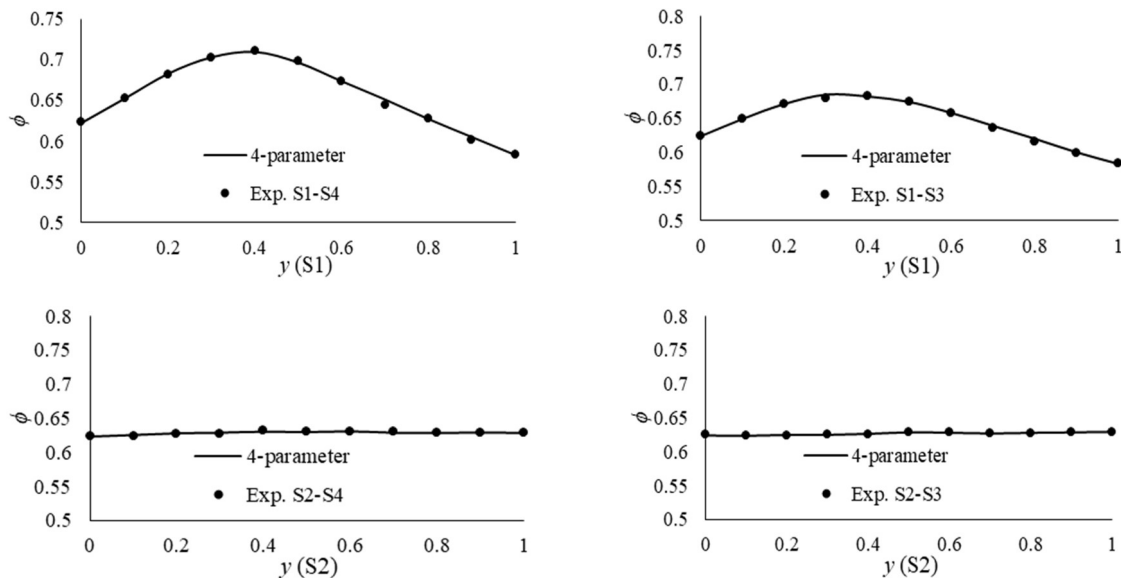

Figure 3. Packing density of mixes S1-S4, S1-S3, S2-S4, S2-S3, S5-S7 and S6-S8.
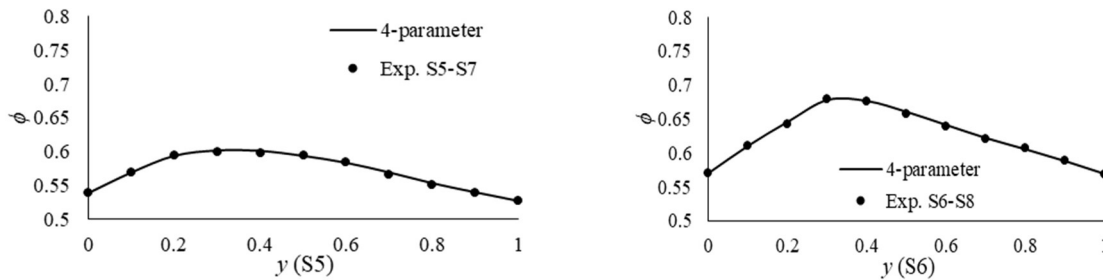
A common characteristic is visible on individual diagrams in figure 3. The significant overestimation of packing density predicted by model is visible in contrast to experimentally determined values. The effect of non-sphericity of particles on packing density was predicted and demonstrated by several authors [9, 10, 11]. Due to the fact that the similar materials are used according to granulometry, as in the previous study, it can be concluded that the deviation is caused by different packing density of individual particle size classes ϕ_i. The value of ϕ_i is independent on particle size and for systems of spherical particles has a constant value of ϕ_C. It seems to be that the deviation from spherical shape of particle causes the decrease of individual particle size class packing density ϕ_i and therefore of whole particular system. Providing an iteration of packing density using calibrated modified 3-parameter model where the value of ϕ_i is replaced by variable results in minimal deviation of modelled and experimentally determined packing density. When the newly determined value of ϕ_i is placed against parameter characterises particle shape, the circularity C^*, following dependence can be obtained (figure 4).

Figure 4. Influence of particle shape on packing density of individual particle size class ϕ_i.

The limit value 0.64 for spherical particle ($C = 1$) is set according [12]. In addition, the samples with C^* in the range of 0.94 to 0.96 are included to the context with determined results of packing density from previous study [8]. It is evident that the packing density of individual particle size class ϕi can be described as a function of particle shape, in this case as standardized circularity C^* and can be fitted by fourth order polynomial equation. Based on the regression curve the value of ϕ_C can be estimated with relatively good precision for particle mixes with standardized circularity of 0.81 to 1 (or equivalent sphericity 0.66–1). To increase model precision the circularity of each particle size class C_i can be defined directly from microscopic image analysis. The parameter C^* can be then directly replaced by C_i in regression equation in figure 4 for direct calculation of ϕi. Implementing these results into the calibrated modified 3-parameter packing model, the fourth parameter dealing with ϕ_i as a function of C_i respectively, following results of packing density can be obtained (figure 5).
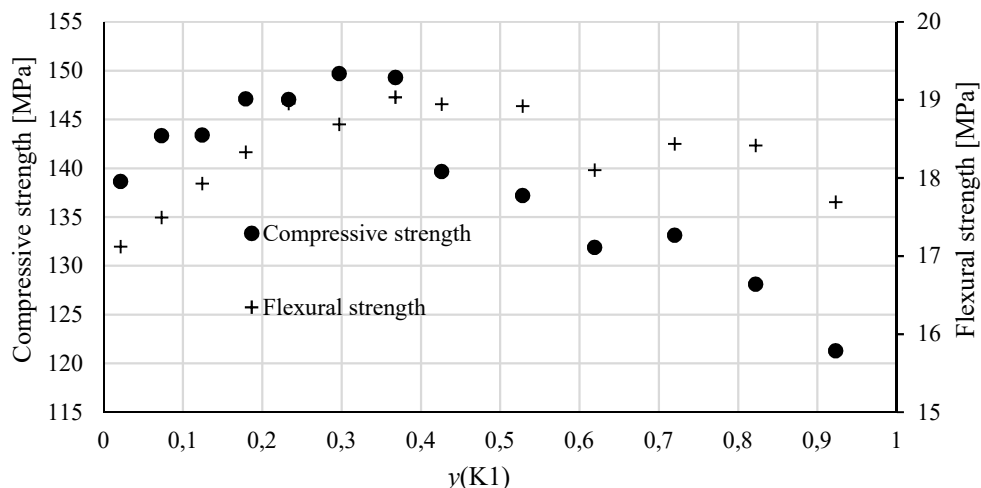
Figure 5. Packing density prediction using 4-parameter packing model.
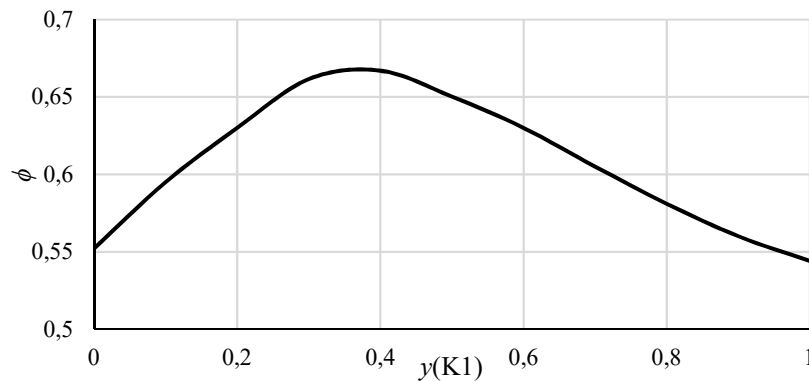
As can be seen there is a very good agreement between experimentally determined packing densities and those obtained by 4-parameter packing model. Calculation via model is suitable for systems with continual granulometry (S2-S4, S2-S3) as well as for systems with gap in particle size density distribution (S1-S4, S5-S7, S6-S8). The suitability of the 4-parameter model is confirmed for particular systems with particle size from approximately 100 µm to 2000 µm. Therefore it can be a very suitable tool in HPC and UHPC design. It is clearly visible that maximal packing density is reached by sample combination with the largest difference in particle size where combination S1-S4 exceeds the packing density of 0.7 at volume content of S1 0.38. On the other hand it seems to be that continual way of aggregate grading does not lead to significant change of packing density as the mixes S2-S4 and S2-S3 show.

The importance of dense packing with maximal density is demonstrated by the results of mechanical properties. The compressive strength and the flexural strength was monitored by samples of UHPC mortars in dependence on aggregate composition i.e. on aggregate packing density. Figure 6 describes the evolution of mechanical properties; the compressive strength and the flexural strength of test specimens of UHPC mortars with variable composition of aggregate skeleton. The aggregate composition is described via volumetric ratio of fine fraction y(K1). The volumetric ratio of fine and coarse aggregate fraction causes the change of aggregate packing density as is described in figure 7.
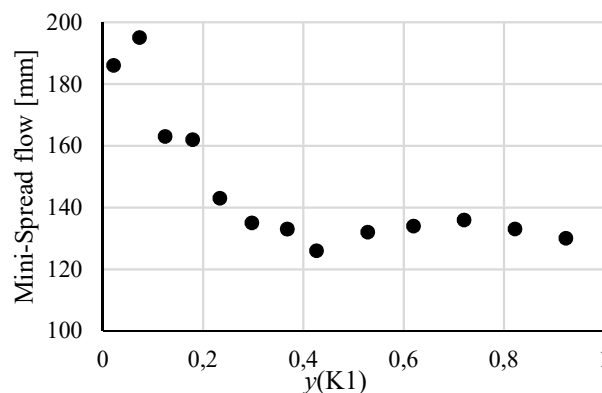

Figure 6. Compressive and flexural strength of prepared samples in dependence on aggregate skeleton composition expressed by volume fraction of fine corundum aggregates y(K1).

Figure 7. Packing density of particular system K1-K2.

It is evident that maximum of mechanical properties is reached at aggregate composition y(K1) and y - 1(K2), respectively with the highest packing density (0.66–0.67) of mix K1-K2. With increasing y(K1) up to 0.35 the mechanical parameters are increasing as well from value of 138 MPa up to value of 150 MPa for compressive strength and from value of 17 MPa up to value of 19 MPa for flexural strength. It is clearly evident that compressive strength is much more affected by packing density than the flexural strength. Moreover, with further increasing of y(K1) the compressive strength drops at the value of 121 MPa when the flexural strength drops at nearly same value as for y(K1) $\approx$ 0. The rapid decrease of the compressive strength is caused not only by reduction of packing density but probably by decrease of workability (spread flow) due to increasing amount of fine aggregate fraction as is visible from figure 8.

Figure 8. Workability via Mini-spread flow test of fresh samples depending on volumetric fraction of K1 in aggregate skeleton.

Similar results can be found in [13]. The workability decreases with increasing volume fraction of K1 to approximately constant level with the local minimum at maximal packing density of mixture K1 and K2 i.e. y(K1) = 0.35. It is believed that low workability for y(K1) in range 0.35–1 is partially responsible for decrease of compressive strength. The spread flow is a consequence of the yield stress changes in fresh mortar. Whereas the yield stress is exceeded by vibration during compaction, there has to be an increase in plastic viscosity of mortar as well. The flow behaviour of mortar is driven by this effect after exceeding the yield stress and therefore negatively effects the compaction of material (due to higher value of plastic viscosity).

5. Conclusion

The packing density of particular systems consisting of non-spherical particles was investigated. It was found that there is a significant influence of particle shape on packing density of the system. Packing

density of such a "real" system cannot be predicted by packing models based on discrete packing theory which was experimentally and analytically proven. Therefore, another parameter has to be found which puts particle shape in the context to packing density. It was found that particle circularity determined from microscopy image analysis effects the pacing density of individual particle size classes and it can be implemented to the calibrated modified Kwan's 3-parameter packing model for multimodal particular systems derived in author's previous study as a fourth parameter to the form of 4-parameter packing model. This newly developed model shows very good agreement with experimental results of packing density for multimodal particular systems of non-spherical particles. The practical use of developed model as well as importance of packing density point-of-view in ultra-high cementitious composite design was verified. The packing density of aggregate skeleton has a great influence mainly on compressive strength of ultra-high cementitious composites where the highest value is reached by sample with the highest packed aggregates. Moreover, increased packing density of aggregates affects the behaviour of material in fresh state due to reduction of binder volume necessary for system lubrication. It is evident that the packing density represents an approach which is necessary to be used in ultra-high performance concrete and other engineered cementitious composite design.

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