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RANDOM DENSE PACKING OF CONTINUOUS DISTRIBUTIONS OF SPHERICAL PARTICLES – USE OF PARTICLE PACKING MODEL AND DISCRETE-PARTICLE-METHOD TO PREDICT PARTICLE SPACING FACTORS

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Abstract. Portland Cement (PC) is responsible for about 8% of global CO2 emissions and is mainly used as a binder in concrete, which is a basic material for all types of load-bearing structures. Due to its versatile usage (precast or ready-mix concrete) and availability almost all over the world, concrete is used in a wide range of applications. When looking at possible pathways to reduce the carbon footprint four measures emerge:

1. Use of renewable energy for PC production (mainly clinker burning process)
2. CO_2 capture during PC production.
3. Use of supplementary cementitious materials to reduce the amount of PC.
4. Optimization of PC efficiency within the concrete to reduce the amount of PC needed.

To optimize the efficiency of PC, particle packing methods can be used, see e.g. [1], [2]. A well-established geometrical particle packing model is the compressible packing model (CPM) [3], which was extended to the compressible interaction packing model (CIPM) to account for colloidal interactions [4]. Based on the CIPM a so-called cement-spacing-factor (CSF) is calculated [2], [4] by dividing the PC volume in concrete by the maximum packing density (PD). CSF is then used to optimize PC efficiency, i.e., strength of PC based materials. However, CSF calculated based on PD is only an auxiliary parameter (i.e., particle saturation on a volume base), which at best qualitatively describes PC spacing in a particle arrangement and does not take the spread of the PC spacing into account. In this paper, a DEM-based enriched particle packing algorithm is developed that accounts for particle type (i.e., inert particles or PC) and distribution of PC particle distance. The results are compared to strength tests on PC-based pastes and the correlation to the calculated distribution of PC particle distance is shown. Based on PD calculations and the enriched DEM particle packing algorithm an optimization of PC efficiency in concrete can be performed.

Keywords: Portland Cement, CO_2 emissions, compressible packing model, DEM, PC spacing, PC efficiency.

1 INTRODUCTION

Concrete is an indispensable material for modern construction. It is cheap, easy to produce, durable, load-bearing, and the raw materials are available in large quantities. However, concrete contributes to the climate crisis, accounting for 8% of global carbon emissions. The main constituents in concrete are water, mineral aggregates and Portland Cement (PC). The latter is mainly responsible for the high carbon dioxide footprint of concrete. In order to reduce the global warming potential of concrete, materials research focuses on the replacement of PC and the reduction of PC content in concrete.

To find a reasonable amount of cement per cubic meter of concrete, the findings according to Adams [1] or Walz [2], are usually applied. These findings link the water-to-cement ratio (w/c-ratio) in the mixture to the compressive strength (CS) of the concrete. However, the CS is a multivariate characteristic parameter of concrete. In addition to the w/c-ratio, the CS is also influenced by the packing density (PD) and the Interfacial Transition Zone (ITZ). ITZ is the region around the aggregates with a locally high porosity, due to the wall effect in particle packing and water adsorption at aggregate surface.

To further reduce the amount of PC in concrete, the PD of the concrete can be increased by selecting suitable coarse aggregates and fine aggregates as well as fine powders. To calculate the PD of concrete materials, the Compressive Packing Model (CPM) was developed by de Larrad [3].

Fennis [4] extended the CPM to the Compaction-Interaction Packing Model (CIPM) and proposed to take a closer look at the particle spacing, rather than the w/c-ratio. A calculation method for the particle spacing between PC particles, the so-called cement spacing factor (CSF) was developed and a correlation between CSF and CS was found. However, when Haist [5] applied the CIPM and CSF in his research, he found different correlations between CSF and CS, which also depends on the type of PC. This highlights the need for research and new approaches when packing density, particle spacing, and the correlation to CS are applied for the optimization of concrete. Therefore, in this study, a discrete element method (DEM) was developed to calculate the absolute distance between PC particles and evaluated with experimentally determined CS of different cement pastes.

2 METHODICAL PROCEDURES

Concrete and mortar CS depends on numerous parameters, making it challenging to isolate a single variable for study. This study investigates two series of cement pastes to assess the influence of particle spacing on CS. In the first series, a constant w/c ratio was maintained while incrementally reducing the PC content, thereby increasing the distance between PC particles. The second series maintained a constant PD as far as possible while reducing the PC content, resulting in varying w/c ratios. CS according to DIN EN 196-1, air content according to DIN EN 12350-7, and PD according to Lowke et al. [6] were measured for both series. A Python-based DEM algorithm was developed to calculate the distance between PC particles. This algorithm uses granulometry data of raw materials as well as the experimentally measured PD to determine particle sizes and their quantities. Within this algorithm, particles are randomly distributed within a defined container, with PC and non-PC particles designated accordingly. Once the particles are situated within the container, distance calculations are carried out.

Specifically, the distances between each PC particle and its 100 nearest PC particle neighbors are calculated, to obtain distance values unaffected by the size of the container size.

3 CPM - PACKING DENSITY

In this paper, the CPM by de Larrad [3] is used to calculate the PD of the dry mixtures. First, the virtual PD γ is calculated. The virtual PD is defined as the maximum PD of a given mixture, with given particle sizes and given self-packing density. The compaction index K is introduced to calculate the real packing density Φ. The compaction index K depends only on the applied compaction and was determined empirically for different types of compactions. The real packing density is then determined indirectly from the compaction index. The PD calculated with the CPM is therefore only dependent on the particle sizes of the dry mix, the associated inherent packing densities and volume fractions, and the type of compaction, and does not take into account the type of particles (e.g. whether they are PC particles or limestone powder particles). The generalized equation to calculate the virtual PD γ for n classes of grains is expressed in Eq. (1)

$$\gamma = \frac{\beta_i}{1 - \sum_{j=1}^{i-1}[1 - \beta_i + b_{ij}\beta_i(1 - 1/\beta_i)]\, y_j - \sum_{j=i+1}^{n}\left[1 - \frac{a_{ij}\beta_i}{\beta_i}\right] y_j} \tag{1}$$

The virtual PD is the maximum PD achievable if the particles were placed one by one and each particle keeping its original shape, therefore it is by definition non-accessible by experiments. In this equation β_i represents the self-packing density, a_{ij} the loosening and b_{ij} the wall-effect. The volume fraction of the dominant class is represented as y_i. The coefficients a_{ij} and b_{ij} were determined empirically by de Larrad and are calculated by equations 2 and 3.

$$a_{ij} = \sqrt{1 - (1 - d_j/d_i)^{1.02}} \tag{2}$$

$$b_{ij} = 1 - \left(d_i/d_j\right)^{1.50} \tag{3}$$

The compaction index was also determined empirically by de Larrad, in this paper k=9 was chosen. The real packing density Φ is determined indirectly from Eq. (4)

$$k_i = \sum \frac{y_i / \beta_i}{1 / \Phi - 1 / \gamma_i} \tag{4}$$

In these investigations a MATLAB CPM script was developed, that combines different materials and calculates the real packing density Φ for every useful combination.

4 EXPERIMENTAL PACKING DENSITY

The experiment after Lowke et al. [6] for measuring the packing density of cement and additions was used to determine the PD of the investigated cement pastes. Roughly 120 ml paste was poured into a cylindrical extraction device and compacted with a defined pressure until the compaction rate was lower than 0.002 mm/min, so equilibrium was reached. Excess pore solution was expressed and collected during the compaction process through a filter at the bottom of the extraction device. This was carried out with pressures 2, 4, 10, and 40 MPa. The volume of the pore solution remaining in the suspension corresponds to the volume of the voids in the particle mixture for the pressure applied. The void content after the compression was determined by drying the compacted sample at 105 °C until mass constancy was achieved. The experimental PD $\phi_{paste,exp}$ was obtained by fitting the data and extrapolating back to zero using quadratic regression.

5 MATERIALS USED

The cement used is a Portland Cement CEM I 52,5 R SP, a cement with a high specific surface, i.e. a high fineness and a minimum of 95% PC. In order to reach a high PD of the pastes, with the possibility to incrementally decrease the PC content and keeping the PD constant, PC with high fineness and three different limestone powders (Betocarb HP-GU, KSM Minerals Conf, Fueller Z) and one quartz flour (Quartz flour W3) with different granulometries are used as raw materials, see Fig. 1. The granulometries were measured using a laser particle size analyzer.

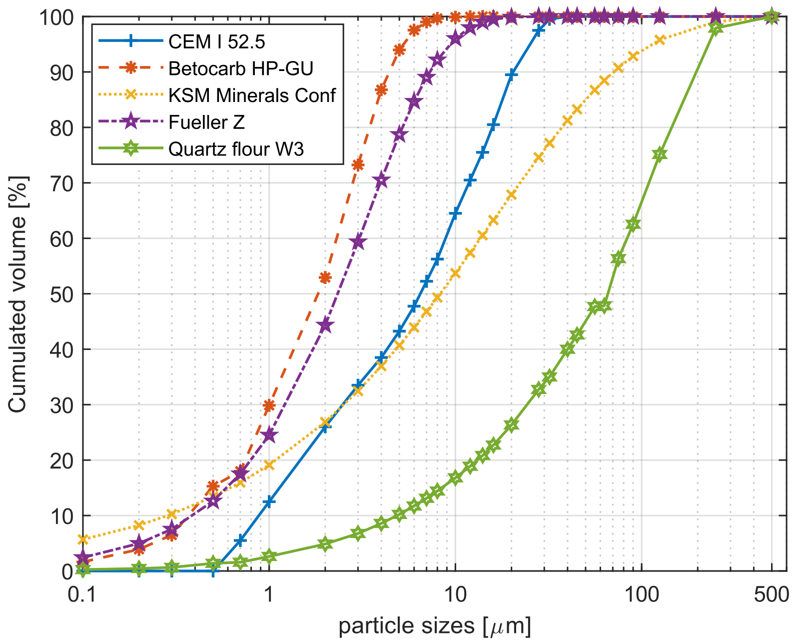


Figure 1: Granulometries of the materials used

In order to ensure accurate and correct weights when producing the cement pastes, the density of the raw materials was measured with a gas pycnometer, and the mean value was calculated from four individual values; the results are shown in Table 1.

Table 1: Example of the construction of one table

Material	CEM I 52.5	Betocarb HP-GU	KSM Minerals Conf	Fueller Z	Quartz flour W3
Density [g/cm³]	3.22	2.84	2.85	2.81	2.69

Additionally, different amounts of the superplasticizer (SP) were used to reach similar workability on every paste with changing water content

6 INVESTIGATED PASTES

This study investigates two cement pastes series to understand the effects of particle spacing on CS. In the first series, PC content is incrementally reduced while maintaining a constant w/c ratio, see Tab. 2. In the second series, the PC content is similarly reduced, but the water content is kept constant, with a granulometric composition so that the PD is as constant as possible, see Tab. 3.

Table 2: test series one

	Solid PC fraction [%]	Volume PC [dm/m³]	Mass PC [kg/m³]	Mass Water [kg/m³]	w/c-ratio [-]
M_C28_0.60	28	179.30	577.71	344.64	0.597
M_C26_0.60	26	170.77	550.21	328.20	0.597
M_C22_0.60	22	152.30	490.71	292.73	0.597
M_C20_060	20	142.30	458.49	273.50	0.597

Table 3: Test series two

	Solid PC fraction [%]	Volume PC [dm/m³]	Mass PC [kg/m³]	Mass Water [kg/m³]	w/c-ratio [-]
M_C28_0.60	28	179.30	577.71	344.64	0.597
M_C26_0.64	26	166.51	536.51	344.56	0.642
M_C22_0.76	22	140.90	453.97	344.56	0.759
M_C20_0.84	20	128.09	412.70	344.56	0.835

7 DISCRETE ELEMENT METHOD (DEM)

A Python-based DEM algorithm was developed to calculate the distance between the PC particles in the investigated cement pastes. In the first step, the algorithm calculates the appropriate mixing ratio based on the stored granulometry of the raw materials and the PD and thus calculates the number of particles of each particle size. The stored granulometry needs to be limited, in order to get a finite number of particles, in this study the lower bound of 6 µm was chosen, the stored granulometry is visualized in Figure 2.

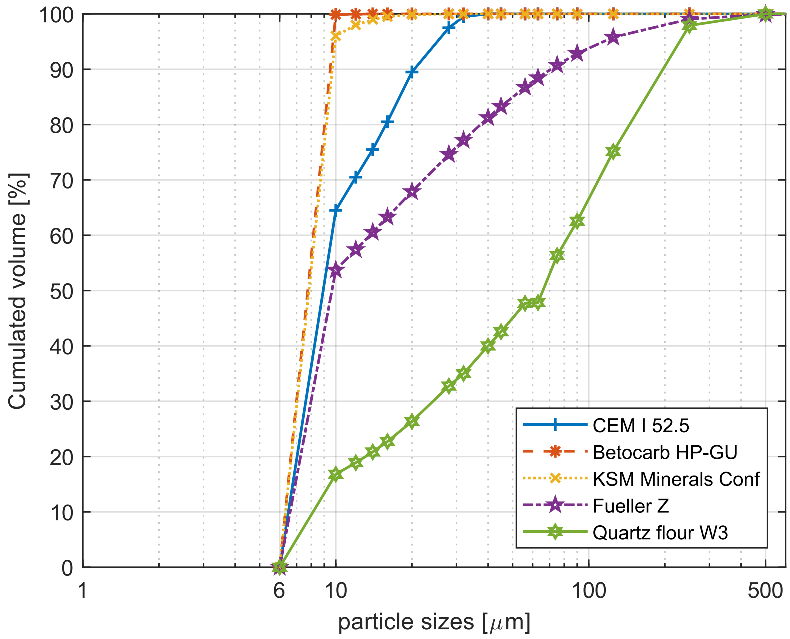


Figure 2: Input Granulometries of the materials used for the DEM algorithm

A container, in which the particles are placed, also needs to be defined, here a cuboid with an edge length twice the diameter of the largest particle – 2 * 500 µm – was chosen. The placing of the particles is not grid-based. Particles are placed as spheres one by one, starting with the biggest particles, by generating a randomized position in µm -resolution for the center point of the sphere. The random function only generates positions that lie within the container and have a minimum distance to each container edge of the radius of the placed sphere. In the next step, a box is created with the center on the previously generated coordinates and an edge length of the sum of the radii of the currently placed particle and that of the largest placed particle. Inside this box is every particle with which the new particle to be placed could intersect. Then, an overlap-function checks if the distance between the generated center point and all existing center points in the box is greater than the sum of radii of the generated and the existing particles. If the distance is less, the randomizing function generates a new center point until the distance requirement is met. This method is not computationally intensive, therefore very fast, and allows RAM-friendly storage of all existing particles, as only the center and radius of each sphere need to be stored. After all particles are placed, the particles are binary classified as PC (‘1’) or non-PC (‘0’). The absolute number of PC particles of each size is calculated, and a random function selects the appropriate particles and assigns them a one, the remaining particles are assigned a zero value. Subsequently, the distance from each PC particle to the 100 nearest PC particle neighbors is calculated, to obtain distances that are as unaffected as possible by the size of the container. From these distance values, the mean value and the standard deviation are calculated.

8 RESULTS AND INTERPRETATION

The data collected are listed in Table 4. The fresh concrete bulk density as well as the air content was determined in a one-liter air-content test device. The hardened concrete density as well as the CS was measured eleven days after manufacture on three prisms each, in Table 4 the mean values are shown.

Table 4: data collected

	fresh concrete density [kg/m³]	air content[%]	$\phi_{paste,exp}$	CS [MPa]	100NN distance
M_C28_0.60	2106	1.5	0.737	68.19	26.67
M_C26_0.60	2124	1.4	0.788	62.78	26.73
M_C22_0.60	2060	1.3	0.836	62.35	27.72
M_C20_0.60	2046	2.2	0.832	56.53	28.68
M_C26_0.64	2038	1.7	0.746	68.05	27.24
M_C22_076	2100	1.1	0.763	50.66	28.60
M_C20_0.83	2108	0.6	0.789	56.80	29.20

8.1 Packing Density

Experimental results indicate an increase in packing density with higher superplasticizer content. This can be attributed to the mode of action of the superplasticizer, which sterically repels PC particles and thus prevents agglomeration of the PC particles. Therefore, a higher packing density of the PC particles can occur. A correlation with an R^2 of 0.795 between superplasticizer content and packing density is observed, see Fig.3.

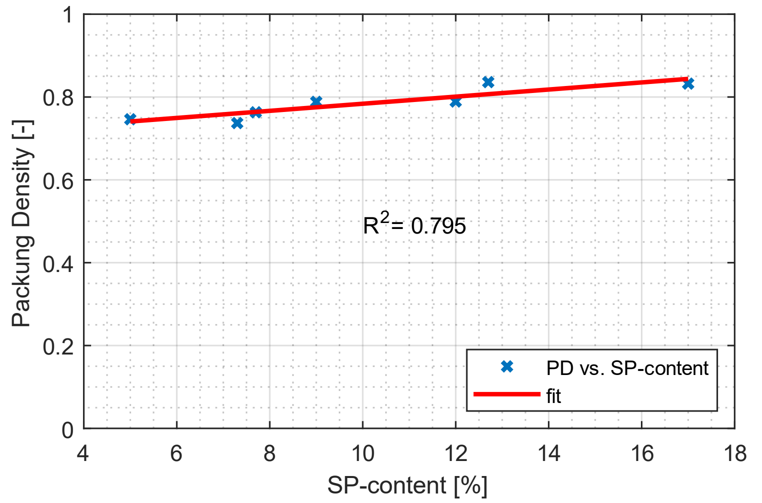


Figure 3: Correlation of PD and SP-content 1

8.2 Compressive Strength

The mean CS after eleven days, which was calculated from six individual values and the corresponding standard are shown in Figure 4.

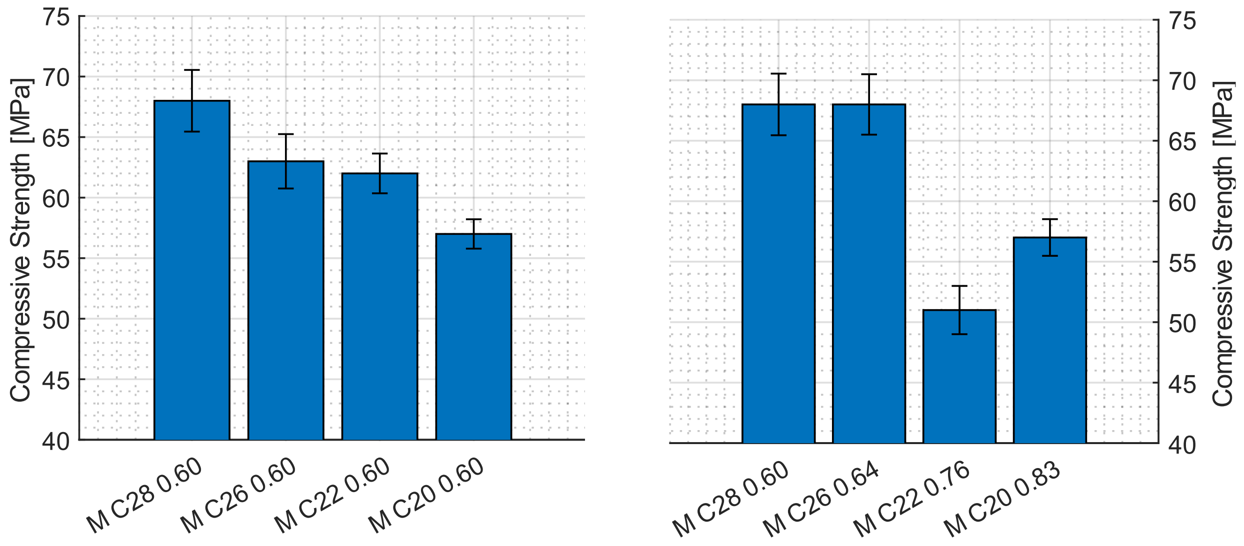


Figure 4a and b: Compressive Strength after 11 days

In the first series, the CS is decreasing with decreasing PC content. It would have been expected that the CS of series 2 would decrease significantly more than that of series 1 as the PC content was reduced incrementally while simultaneously lowering the w/c-ratio. Figure 5 visualizes the observed correlation between CS and w/c-ratio with an R^2 of 0.360. This contradicts with every law known for design of concrete composition like the ones of Adams [1] or Walz [2].

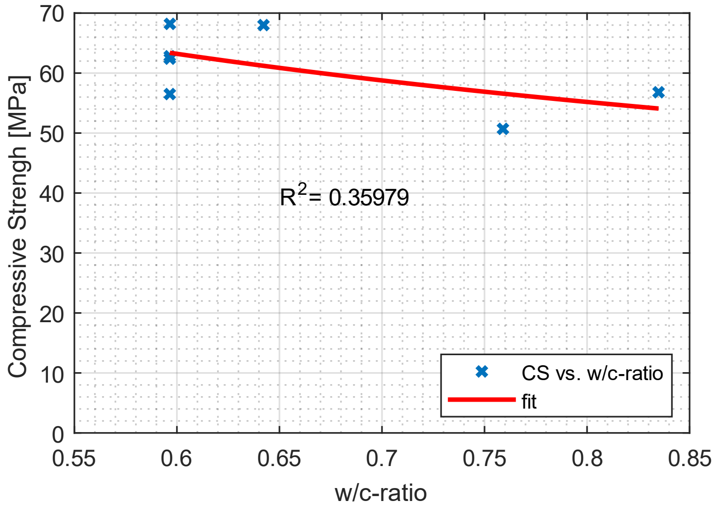


Figure 5: Correlation of CS and w/c-ratio

8.3 Particle Spacing

The average distances as well as the standard variance, obtained by the DEM simulation are shown in Fig 6.

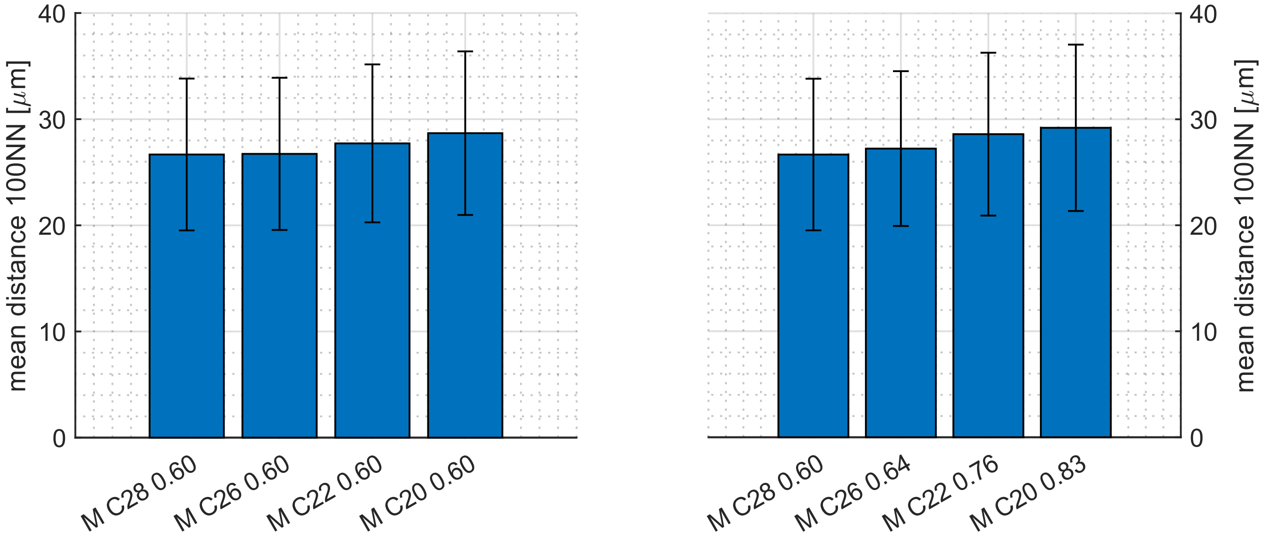
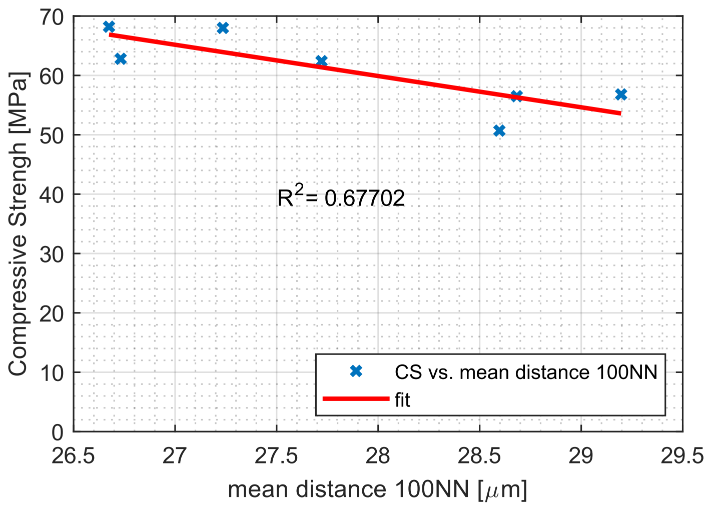


Figure 6a and b: mean PC distances

Notably, the standard deviations should be viewed as indicative of the range of distances among the 100 nearest neighbors, offering a comprehensive view of the distribution. Of particular interest is the consistent pattern observed in the mean distances. Significantly, as the PC content decreases, there is a corresponding increase in PC-particle distances. Similarly, higher w/c-ratios lead to a greater separation between PC particles. These observations align with expectations and underscore the influence of PC content and w/c ratios on particle spacing. A notable finding emerging from this analysis is the correlation between PC-spacing and CS. This correlation is supported by an R^2 value of 0.677, as depicted in Figure 7.

Figure 7: Correlation of CS and PC distances



9 CONCLUSIONS

In this study, to evaluate the influence of particle spacing on CS, two series of cementitious pastes were investigated. CS, air content and packing density were experimentally measured. In addition, particle spacing was quantified, by calculating the distance between PC particles using a DEM algorithm. This algorithm relies on granulometry data from raw materials as well as the PD to ascertain both the sizes and quantities of the particles. Within the algorithm, a container is defined into which the particles are placed serially, one by one. The closest 100 PC particles of each PC particle are found, and their distances are calculated. From these distances, the mean value and the standard deviation were calculated. The results demonstrate a stronger correlation between PC distances and CS compared to the w/c ratio and CS. This underlines the importance of particle spacing for the optimization of concrete and mortar mixes to improve CS with lower amounts of PC. However, there is still a great need for research and improvement of the DEM algorithm. The serial DEM algorithm took roughly 12 to 16 hours to place about $2 \cdot 10^6$. particles. Therefore, a parallelization of the process is to be strived for to allow for an optimization algorithm. In addition, quantification of the PC spacing in the form of distance calculation of 100 neighbors is the first idea that should be developed through further research. The predictability of this quantification becomes uncertain and may prove unsuitable when alternative container sizes and compositions, such as the inclusion of coarse aggregates and different types of cement, are introduced. Furthermore, ITZ, as one of the most important factors influencing CS, which in future should be addressed within in the DEM simulation.

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