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Spatial dispersion of aggregate in concrete a computer simulation study

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Abstract. Experimental research revealed that the spatial dispersion of aggregate grains exerts pronounced influences on the mechanical and durability properties of concrete. Therefore, insight into this phenomenon is of paramount importance. Experimental approaches do not provide direct access to three-dimensional spacing information in concrete, however. Contrarily, simulation approaches are mostly deficient in generating packing systems of aggregate grains with sufficient density. This paper therefore employs a dynamic simulation system (with the acronym SPACE), allowing the generation of dense random packing of grains, representative for concrete aggregates. This paper studies by means of SPACE packing structures of aggregates with a Fuller type of size distribution, generally accepted as a suitable approximation for actual aggregate systems. Mean free spacing $\overline{\lambda}$, mean nearest neighbour distance (NND) between grain centres $\overline{\Delta_3}$, and the probability density function of Δ_3 are used to characterize the spatial dispersion of aggregate grains in model concretes. Influences on these spacing parameters are studied of volume fraction and the size range of aggregate grains. The values of these descriptors are estimated by means of stereological tools, whereupon the calculation results are compared with measurements. The simulation results indicate that the size range of aggregate grains has a more pronounced influence on the spacing parameters than exerted by the volume fraction of aggregate. At relatively high volume density of aggregates, as met in the present cases, theoretical and experimental values are found quite similar. The mean free spacing is known to be independent of the actual dispersion characteristics (Underwood 1968); it is a structural parameter governed by material composition. Moreover, scatter of the mean free spacing among the serial sections of the model concrete in the simulation study is relatively small, demonstrating the sample size to be representative for composition homogeneity of aggregate grains. The distribution of Δ_3 observed in this study is markedly skew, indicating a concentration of relatively small values of Δ_3. The estimate of the size of the representative volume element (RVE) for configuration homogeneity based on NND exceeds by one order of magnitude the estimate for structure-insensitive properties. This is in accordance with predictions of Brown (1965) for composition and configuration homogeneity (corresponding to structure-insensitive and structure-sensitive properties) of conglomerates.

Keywords: aggregates; computer-simulation; dispersion; SPACE; spacing, stereology.

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1. Introduction

Properties and the spatial dispersion of the aggregate exert significant influences on the mechanical and durability properties of concrete. With respect to mechanical performance, individual aggregate grains stimulate interfacial crack initiation and crack propagation, but also act as crack arresters (Stroeven, P. 1974). On the other hand, a relatively high internal porosity in the interfacial transition zone (ITZ) between the aggregate surface and bulk cement paste is a relevant phenomenon in durability studies. Simulation studies by Chen (2003) indicated that a smaller spacing between aggregate grains leads to higher porosity connectivity in the paste pockets between aggregates grains. Insight into the spatial dispersion (i.e. spacing between aggregate grains) is therefore of paramount importance for mechanical as well as durability performance. Stereology offers spacing parameters for characterization of the spatial distribution of particles. Stroeven investigated the global interdependence of aggregate and crack structures with the help of stereological techniques (Stroeven, P. 1973, 1974). In his experimental research, the mean free spacing and average nearest neighbour distance between aggregate grains were deduced from section images of serially sectioned concrete specimens. It is generally not possible to directly extract three-dimensional information on spacing in concrete structure by way of experimental methods, unless serial sectioning is accomplished. The exception is the mean free spacing that is the same in a section as in space. Apart from such inherent limitations in its capabilities in yielding spatial information on materials structure, experimental approaches are time-consuming, and thus expensive. Hence, computer simulation offers an alternative to at least partly overcome these deficiencies.

The volume fraction of aggregate in ordinary concrete varies between 60~80%. Conventional simulation approaches based on random-generator procedures are not able to generate packing systems of aggregate grains with such densities. In this paper, use is therefore made of the SPACE (Software Package for the Assessment of Compositional Evolution) system for generating structures of densely packed aggregate grains on the basis of a Fuller sieve curve. Mean free spacing, $\overline{\lambda}$ mean NND between aggregate grain centres $\overline{\Delta_3}$, and the probability density distribution of Δ_3 are used to represent the spatial dispersion of aggregate grains in model concretes. The investigated cases cover a moderate range of volume fractions and maximum diameters of aggregate grains with respect to container size. The influences of these technical parameters are studied on the aforementioned spacing descriptors. Measurement of these stereological parameters is readily accomplished by the SPACE system. Comparison between experimental results and predictions (based on uniformly random or completely ordered structures) can offer insight into the degree of order of aggregate structures in the investigated concrete mixtures, and allows assessing the configuration-sensitivity of the spacing parameters.

Every independent geometrical parameter has its own scale of homogeneity, which can be associated with the corresponding representative volume/area element (RVE/RAE). Definitions for the RVE range from the least restrictive one of composition homogeneity to the other extreme based on configuration homogeneity (Stroeven, P. 1973, Freudenthal 1950). Hence, structure-insensitive and structure-sensitive properties of concrete can be associated with different geometric parameters of material structure, and require RVE's of quite different sizes for their global assessment.

2. Material designs and modelling

The concrete structure can be considered for this purpose as a two-phase system, i.e., aggregate grains dispersed in a uniform matrix. The aggregate is assumed composed of spherical grains of which size complies with the Fuller distribution function: $G(d)=(d/d_m)^{1/2}$, where $G(d)$ is the volume-based percentage of particles with diameters smaller than d. The minimum and maximum diameters of aggregate grains are denoted as d_0 and d_m, respectively. The Fuller distribution function is generally accepted as a suitable approximation of actual aggregates system in concrete, whereas sphericity is considered a suitable approximation (reducing computing time) for aggregates of fluvial origin. The minimum diameter is set at 1 mm for all investigated mixtures. Model concretes were generated with three values of d_m, i.e., 15, 20 and 32 mm, respectively. Three volume fractions of aggregate grains (V_V) were involved in this study, i.e., 60%, 65% and 70%. Initially, the aggregate grains were located by random generator system inside a sufficiently large cubic container with periodic boundaries, to represent bulk material conditions. The grains were then subjected to a dynamic mixing process, while the container size was gradually reduced until a desired volume fraction of aggregate was achieved. A parameter in the SPACE system accounts for the energy loss due to internal friction between the aggregate grains and the matrix material. So, the obtained model material represents the fresh state of concrete. For a detailed description of the SPACE system and relevant algorithms, see (Stroeven, P. & Stroeven, M. 1999, Stroeven, P. 2000a). Care should be bestowed on the container size *after compaction* in view of the size of the representative volume element (RVE) for the geometric parameter at issue. This is inevitably a compromise, since increase in container size is accompanied by a disproportional increase in computing time. Total sample could encompass a series of simulations based on repeated SPACE-generations of similar aggregate structures. Although average parameter results obtained on sub-RVE samples are biased estimators of associated global values of the respective RVEs (when structure-sensitive property is involved), averaging among repetitions is a sensible approach for comparison purposes *on the same sampling resolution level*. Moreover, the sample repetitions could be *added* to get eventually to the representative sampling level.

Concrete belongs to the category of macroscopically heterogeneous materials. This implies RVEs to have macroscopic dimensions. The linear dimensions of the RVE will range for concrete from, say, 100 to 1000 mm, depending on the structural or mechanical property of interest (Stroeven, P. 1974). According to the rule of thumb in concrete technology, the smallest linear dimension of the container should be at least four times the maximum diameter of aggregate grains, d_m. However, this rule is derived from estimates of Young's modulus based on strain gauge measurements (Cooke and Seddon 1956), so deals with a rather structure-insensitive property. For practical values of d_m, we would indeed end up at the bottom side of the aforementioned practical range of linear dimensions of the RVE. Container size in the simulation program is mostly of representative size as far as configuration-insensitive parameters are at issue.

3. Spacing parameters

3.1. Definitions of spacing parameters

Quantitative stereology provides the tools for three-dimensional characterization of the aggregate dispersion in a cementitious matrix. The frequently used stereological parameters for assessing

spacing between particles are the free spacing, λ, and the nearest neighbour distance Δ_3. Mean free spacing $\overline{\lambda}$ represents the uninterrupted surface-to-surface distance to all possible neighbours averaged for all particles, as opposed to $\overline{\Delta_3}$ that expresses an average value of inter-particle distances between nearest neighbour centres only. Since average values of these parameters are important quantities when considering bulk properties of concrete, mean free spacing $\overline{\lambda}$, and mean nearest neighbour distance $\overline{\Delta_3}$ are measured. Further, the mean intercept length through the particles, $\overline{L}_3$, is the mean value of particle intercepts with a randomly positioned and oriented sweeping test line system; as a consequence, it characterizes particle size. Still, it connects two spacing parameters, the mean free spacing $\overline{\lambda}$ and the *mean particle spacing, L_V,* by $\overline{L}_3 = L_V - \overline{\lambda}$. Worthy of note is that the mean intercept length through the matrix, $\overline{L}_m$, equals $\overline{\lambda}$, whether measured in the section plane or in space. Of course, $\overline{L}_3 + \overline{L}_m = L$, the total test line length. The definitions and expressions of these stereological descriptors can be found in Underwood (1968). The parameters can be calculated as follows (Hu and Stroeven 2003):

$$\overline{L}_3 = 4\frac{V_V}{S_V} = 4\frac{N_V \frac{1}{6}\pi \overline{d^3}}{N_V \pi \overline{d^2}} = \frac{2\overline{d^3}}{3\overline{d^2}} \tag{1}$$

$$\overline{\lambda} = 4\frac{1-V_V}{S_V} = \overline{L}_3 \frac{1-V_V}{V_V} = \overline{L}_m \tag{2}$$

where N_V is the number of particles per unit of test volume, V_V is the volume fraction of aggregate, S_V is the surface area of particles per unit of test volume, and $\overline{d^3}$ and $\overline{d^2}$ are the third and second moments, respectively, of the particle size distribution function (PSD). For the Fuller distribution, $\overline{d^2} = 5d_0^2$ and $\overline{d^3} = 5d_0^{2.5} d_m^{0.5}$ (Stroeven, P. 2000a, Hu and Stroeven, P. 2003). $\overline{\Delta_3}$ is defined for a *uniformly random distribution of point particles* in a test volume (Underwood 1968):

$$\overline{\Delta}_3 = 0.554\frac{1}{\sqrt[3]{N_V}} = 0.554\sqrt[3]{\frac{\pi}{6V_V}\overline{d^3}} = 0.446\sqrt[3]{\frac{\overline{d^3}}{V_V}} \tag{3}$$

3.2. Algorithm for determining spacing parameters

These spacing parameters can be automatically measured by the SPACE system. A two-dimensional scheme for measuring separate values of the free spacing λ_i for a two-phase composite is presented in Fig. 1. It involves superposition in random direction of parallel test lines on the sample section; all unobstructed distances between particle surfaces are recorded and averaged to yield $\overline{\lambda}$. Fig. 2 shows the algorithm for determining Δ_3 by two different methods. Fig. 2(a) is based on a so-called brute force method. The measurement is rather straightforward, since the distances between a target particle and all other particles in the sample are calculated and the minimum value is chosen as Δ_3 for this target particle. Contrary to this time-consuming method, a cell partitioning method (Fig. 2b) is employed in the SPACE system. The sample cube is partitioned into cells of the same size. All particles with their centre point inside a certain cell are considered belonging to this master cell. For a certain target particle, only a limited number of particles located in the master cell and in its nearest neighbour cells should be considered. This method largely reduces computing time. For details of this algorithm, see Stroeven, P. (1999). To prevent biases due to boundary problems, all particles intersected by the orthogonal set of co-ordinate axes are included, whereas all

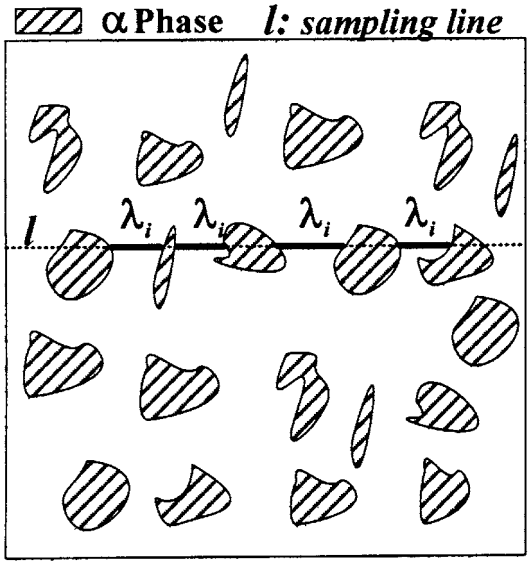


Fig. 1 Schematic of free spacing measurement between dispersed particles (α-phase) for two-phase composites; in this study, the phase of interest is assumedly spherical aggregate grains.

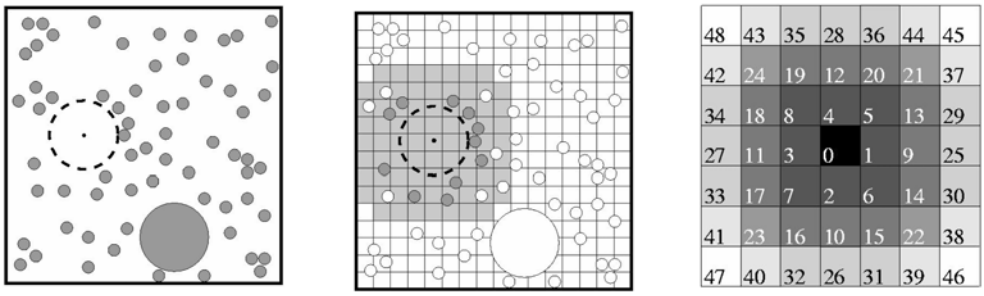


Fig 2 Number of investigated particles (dark grey) for the evaluation of nearest neighbouring distance for a specific target particle (dashed line) by two different algorithms: (a) brute force method, 64 particles and (b) cell partition method, the target cell (numbered as 0) including 11 particles; picture (c) is the schematic for coding of neighbouring partition cells in the calculation (Stroeven, P. 1999).

particles intersected by the other set of orthogonal boundaries are excluded from the procedure.

4. Results and discussions

4.1. Influence of volume fraction and size range of aggregate grains

The values of the spacing parameters for concretes made with aggregate systems of different values of V_V and d_m are given in Table 1. The calculation results based on Eqs. (1)-(3) are compared with direct measurements. The decline in $\overline{\Delta_3}$ as well as in $\overline{\lambda}$ with increasing V_V for a given value of d_m, and the increase of these spacing parameters with coarsening of the aggregate system for a

Table 1 Theoretical and experimental values of spacing parameters

Size range (mm)	V_V (%)	$\overline{\Delta}_{3S}$ (mm)	Centre-to-centre $\overline{\Delta}_3$ (mm)			$\overline{\lambda}$ (mm)	
			$\overline{\Delta}_{3th}$	$\overline{\Delta}_{3ex}$	$\overline{\Delta}_{3ex}/\overline{\Delta}_{3th}$	$\overline{\lambda}_{th}$	$\overline{\lambda}_{ex}$
1~15	60	0.067	1.42	1.62	1.14	1.72	1.80
	65	0.044	1.38	1.57	1.13	1.39	1.44
	70	0.013	1.35	1.52	1.12	1.11	1.14
1~20	60	0.078	1.49	1.64	1.10	1.99	2.05
	65	0.055	1.45	1.59	1.09	1.60	1.66
	70	0.013	1.42	1.55	1.09	1.28	1.29
1~32	60	0.126	1.61	1.74	1.08	2.51	2.71
	65	0.079	1.57	1.67	1.06	2.03	2.17
	70	0.041	1.53	1.61	1.05	1.62	1.74

Note: Subscript 'ex' means measurement, 'th' means theoretical calculation. $\overline{\Delta}_{3S}$ is average surface-to-surface NND.

given value of V_V, are in agreement with expectations. The influence of the size range is found more pronounced than that of the volume fraction. At the relatively high volume densities of aggregates as met in the present cases, the theoretical and experimental values of $\overline{\lambda}$ are quite close. The ratio between theoretical and experimental values of $\overline{\lambda}$ is 0.95 (averaged over three systems with different d_m values) at V_V of 60%. This ratio increases slightly as volume fraction reaches 70%. The mean free spacing is known to be independent of the actual dispersion characteristics (Underwood 1968); it is a structural parameter governed by material composition. Moreover, scatter of the mean free spacing among the serial sections of the model concrete in the simulation study is relatively small, demonstrating the sample size to be representative for *composition homogeneity* of aggregate grains.

The size range of aggregates exerts more significant influences on these spacing descriptors than the volume fraction. According to Eqs. (2) and (3), $\overline{\lambda}$ is proportional to $\overline{d^3}/\overline{d^2} = \sqrt{d_0 d_m}$, and also proportional to $(1-V_V)/V_V$. In the investigated cases, V_V varies within a small range (60~70%), much smaller than the range of d_m values. $\overline{\Delta}_3$ increases proportionally to $\sqrt[3]{\overline{d^3}} = \sqrt[3]{5 d_0^{2.5} d_m^{0.5}}$, but decreases with $\sqrt[3]{V_V}$. This explains the minor effects of aggregate volume fraction. On the other hand, the analytical and simulation conditions are different for systems with different values of d_m. $G(d)$ is a continuous function in the analytical approach, whereas in the experiments it reveals increasing discontinuities when approaching d_m. This phenomenon will get more pronounced when d_m increases, because the number of particles is reduced at higher values of d_m. Hence, probability of having a representative aggregate mixture at larger size ranges declines. This is one of the sources for (small) differences between theoretical and experimental values.

Table 1 reveals the measurements of $\overline{\Delta}_3$ in all cases to exceed the calculations, reflecting a partly ordered configuration. The constant 0.554 in Eq. (3) is derived for a random point system, therefore underestimates the nearest neighbour spacing for finite size particles. Table 1 demonstrates the ratio of experimental and theoretical values to be significantly declining with d_m. The number of particles per unit of volume, N_V, is proportional to $1/\sqrt{d_m}$ (Stroeven, P. 2000b). Hence, it is to be expected that the global degree of order in the aggregate dispersion will diminish at declining number of particles. So, analytical and experimental values will be more alike. Table 1 also shows the influence of V_V on the ratio of experimental and theoretical values to be small. Still, N_V is

proportional to V_V, but local state of order is hardly influenced. In fact all spheres that should penetrate each other in order to fulfil the assumption of randomness pull each other apart. Stroeven experimentally studied the spatial distribution of mono-sized 16 mm spherical aggregate grains in concretes with different volume fractions (Stroeven, P. 1973, 1974). He compared the trends in the changes of $\bar{\lambda}$ and $\bar{\Delta}_3$ (as V_V increases) with predictions based on a single-stagger configuration (a completely ordered structure) and concluded that, contrary to $\bar{\lambda}$, $\bar{\Delta}_3$ was governed by the rules of geometry, i.e., reflected a state of *local order* (Stroeven, P. 1974). So, simulation results are supported by these experimentally found trends.

Another characteristic spacing parameter, i.e., surface-to-surface spacing (Δ_{3S}) between nearest neighbouring aggregates is of higher relevance to mechanical and durability properties of concrete than the centre-to-centre spacing. Relevant simulation data are also presented in Table 1. Based on scanning electron microscopy (SEM) observations of specimen sections, Diamond, *et al.* (1982) estimated average surface-to-surface nearest neighbour distances (Δ_{3S}) to vary between 75~100 μm in ordinary concrete. Koenders (1997) calculated $\bar{\Delta}_{3S}$ to be in the 40~100 μm range for an aggregate system complying with the Fuller size distribution. Chen (2003) proved analytically that $\bar{\Delta}_{3S}$ decreases approximately linearly with aggregate volume fraction in a practical range of particle densities. Stroeven, P. and Stroeven, M. (1997) showed that in agreement with definitions, the following relationship between centre-to-centre and surface-to-surface nearest neighbour spacing parameters holds

$$\bar{\Delta}_{3S} = \bar{\Delta}_3 - \bar{d} \tag{4}$$

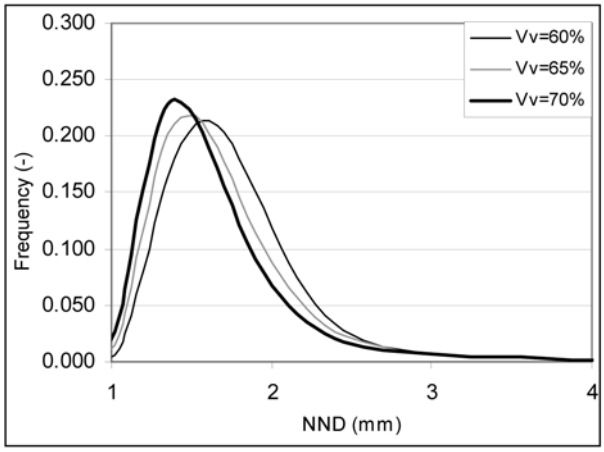
in which $\bar{d} = 5d_0/3$. This is a constant for all mixture; based on the theoretical size distribution curve, $\bar{d}$=1.67 mm. Hence, $\bar{\Delta}_{3S}$ is the difference between two quantities in Eq. (4) of about equal magnitude, as is obvious in Table 1. Hence, with a very small value at maximum packing (V_V around 75% in Stroeven, P. and Stroeven, M. (1999)), $\bar{\Delta}_{3S}$ will increase proportional to $1/\sqrt[3]{V_V}$. Since the aggregate volume fraction range will be small, it can easily be demonstrated that $\bar{\Delta}_{3S}$ is proportional to

$$\left(1 + \frac{1}{3}\frac{\Delta V_V}{\hat{V}_V}\right),$$

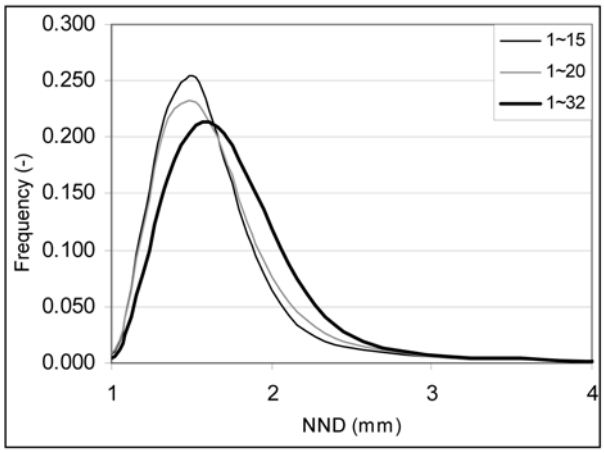
where $\Delta V_V = \hat{V}_V - V_V$, in which $\hat{V}_V$ is the volume fraction at maximum density, and V_V is the actual value of volume fraction of aggregate. Hence, the dependence of the surface-to-surface spacing on aggregate volume fraction is approximately linear in a limited range of packing densities.

4.2. Configuration heterogeneity of aggregate grains

The probability density function of Δ_3 and Δ_{3S} is investigated for all concrete mixtures (Figs. 3 and 4). The trend revealed by computer simulation in Fig. 3 is consistent with experimental observations by Stroeven, P. (1973, 1974). In what follows, the sensitivity of λ, Δ_3 and Δ_{3S} to material configuration will be discussed on the basis of probability distribution of these spacing parameters. The concrete sample was sectioned and the coefficient of variation (CV) of mean free spacing between serial sections is lower than 10% for all investigated mixtures. The relatively small scatter reveals $\bar{\lambda}$ to be relatively configuration-insensitive and, therefore, can be associated with structure-insensitive properties. This is in agreement with the experimental observations of Stroeven,



(a) 1~32 mm aggregate system



(b) $V_V = 60\%$

Fig. 3 Influences of technical parameters (a) volume fraction of aggregate and (b) size range (in mm) of aggregates on probability distribution of centre-to-centre nearest neighbour distance (Δ_3) between aggregate grains.

P. (1974). The representativeness of specimens for studying structure-insensitive properties based on composition homogeneity of the grain structure can be roughly determined on the basis of the ratio of specimen to maximum grain size, accepting a CV for the geometric parameter below 10%. In this study, the ratio of specimen (linear dimension of cubic container) to maximum grain size (d_m) roughly amounted 5 for the 1~15 mm system and 4 for the 1~20 mm system. These size ratios confirm the findings with respect to aggregate composition to be representative.

Contrary to λ, reflecting the uninterrupted distances between all possible pairs of particles, Δ_3 and Δ_{3S} only consider distances between nearest neighbouring pairs of particles. The latter two spacing parameters are sensitive to configuration. Especially Δ_{3S} is expected to be highly sensitive, because it locally measures the result of differences in dispersion as to size and to location. According to

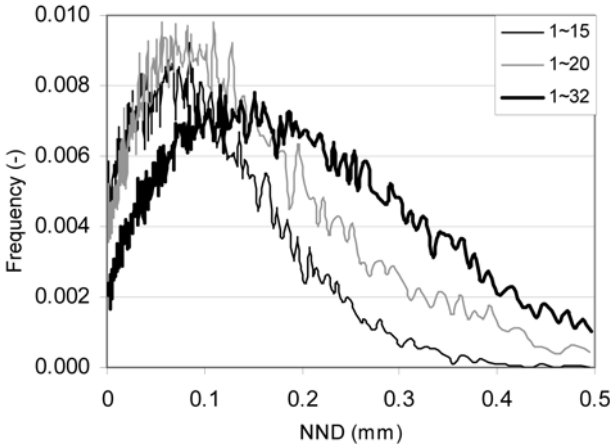


Fig. 4 Probability density distribution of surface-to-surface nearest neighbour distance between aggregate grains for aggregate systems with different particle size range (size range in mm).

Kendall and Moran (1963), the probability density function of Δ_3 for a sample is skew, because $8\pi N_V(\Delta_3)^3/3$ is distributed as χ^2 with 2 degrees of freedom. Fig. 3 indeed reveals a disproportionately large number of relatively small values of the nearest neighbour distance. Apart from allowing the determination of $\overline{\Delta}_3$, this distribution curve offers the possibility to assess the size of the RVE/RAE for configuration homogeneity of the nearest neighbouring spacing among aggregate grains. This is demonstrated for the configuration-sensitive surface-to-surface spacing in section planes, Δ_{2S}. The RVE for this parameter will be relevant for a structure-sensitive mechanical property that would be directly governed by this structural parameter (Stroeven, P. and Stroeven, M. 2001).

All surface-to-surface distances between nearest neighbouring grain pairs are measured on section planes and classified according to their length. The information on the probability density function $f(\Delta_{2S})$ obtained from these measurements is plotted as a histogram with nine length classes. The length range is 0~1.0 mm, with an interval width of 0.1 mm for the first seven classes and 0.15 mm for the last two classes, so that the minimum frequency is at least at the 5% level. Averaging over all section histograms (total number of 40 statistically independent section planes from three orthogonal directions) yields a reference histogram of which the frequency in class i is denoted by $\overline{f}_i$. The corresponding frequency of a single section histogram is indicated by f_i. The χ^2-value is defined by $\chi^2 = \Sigma_{i=1}^{9}(f_i - \overline{f}_i)^2/\overline{f}_i$. Successively adding single sections leads ultimately to an average histogram that sufficiently close resembles the reference one. The accompanying decline in χ^2-value is at that stage arriving at an acceptable limit defining the size of the RAE. The simulation results are presented in Fig. 5 for the 1~32 mm aggregate system (V_V = 60%). The curve in Fig. 5 presents the χ^2-values for the Δ_{2S} histograms at an increasing number of section planes (N) of 100 × 100 mm. These data can be approximated by a smooth curve of hyperbolic type. Global extrapolation of the hyperbolic curve (taking 1.239 as acceptable level, 7 degrees of freedom because frequency of 9th class was lower than 5%) would lead to N=7~9. This implies a RVE for configuration-sensitivity with linear dimension of about 300 mm. The value of $\overline{\Delta}_{2S}$ for this structure is 0.397 mm, i.e., three times the value of $\overline{\Delta}_{3S}$ (0.126 mm). This indicates that nearest neighbours in space are much closer spaced than reflected by the section plane pattern. By using the distribution curve $f(\Delta_{3S})$ for assessing configuration homogeneity, it could be expected that the linear dimension of the RVE will

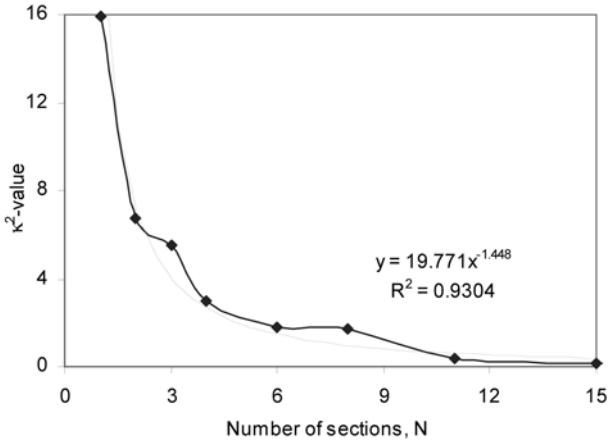


Fig. 5 χ^2-values show differences in $f(\Delta_{2S})$ as revealed by the reference histogram and by a histogram representing an increasing number of section planes, N.

exceed the size of the RVE for structure-insensitive properties as given by the rule of thumb (4 to 5 times maximum structural dimensions) by one order of magnitude. This would be in accordance with predictions of Brown (1965) for composition and configuration homogeneity of conglomerates associated with structure-insensitive and structure-sensitive properties respectively.

4.3. Implications for concrete permeability

The permeability of concrete is closely related to porosity, pore size and tortuosity of transport routes in concrete. Based on the Carmen-Kozeny model, the permeability of concrete (k) can be predicted by:

$$k = \frac{P(V_{pore}/S_{pore})^2}{2\beta} \tag{5}$$

where P stands for porosity, V_{pore} and S_{pore} are the volume and the internal surface area of pore space, respectively, and β is tortuosity of the transport route in concrete. Stroeven, P. (2000b) demonstrated by a stereological approach that tortuosity of the transport route in spherical aggregate concrete can be estimated from a single geometric parameter, i.e., volume fraction of aggregate, $\beta = 1+V_V/2$. Hence, details of the aggregate size distribution will not exert prime influence on transport phenomena of concrete. Volume fraction of aggregate is a composition parameter; therefore, the contribution of aggregate to transport tortuosity in concrete is only related to composition of the material. V_{pore}/S_{pore} characterizes *pore size*. Eq. (2) provides the direct link to the mean free spacing. Since mean free distance can be associated with configuration-insensitivity (Stroeven, P. 1974), concrete permeability can be expected to be structure-insensitive. The simulated diffusion experiments (amount of water transferred over a certain distance through concrete specimen per time unit) by Roelfstra (1989) provide supporting evidences for this conjecture.

Fig. 4 reveals that the major part of Δ_{3S} distribution lies below 0.1 mm, with an extremely high portion even smaller than 0.05 mm. The simulation study by Chen (2002) suggested the threshold surface-to-surface spacing to be 0.069 mm for model concrete with ordinary cement fineness (334

m^2/kg) at water cement (w/c) ratio of 0.3. The interaction (overlap) of interfacial transition zones (ITZs) around neighbouring aggregate grains will become a prevailing phenomenon when the aggregate spacing falls below this threshold value. Under such conditions, porosity connectivity in concrete (i.e. in the ITZ) will increase, as observed (by means of SEM) by Scrivener and Nemati (1995). The value of Δ_{3S} roughly doubled when d_m was changed from 15 to 32 mm at equal volume fraction (Table 1). Hence, a wider size range of aggregate grains leads to favourable results with respect to diminishing ITZ interferences.

The volume fraction of aggregate plays an important role with respect to concrete permeability because the percolation effect of interconnected porosity can be far outweighed by the increasing tortuosity associated with the presence of aggregate grains (Marchand and Delagrave 1999). At higher aggregate volume fraction, tortuosity will increase to a sufficient degree to offset the percolation of porosities in the ITZ. Supporting evidence can be found in the literature (Jaiswal, *et al.* 1997). Jaiswal, *et al.*, experimentally investigating the influence of aggregate volume fraction on concrete permeability, came to the conclusion that the influence of tortuosity (associated with the presence of aggregates) dominated over ITZ's negative effects on permeability, at least up till aggregate volume fractions of about 50%. On the other hand, a slight increase in V_V from 60% to 70% gave rise in our experiments to a significant decline of Δ_{3S} (almost dropped to 1/3 of the initial value). Hence, volume fraction should not exceed 60% for the purpose of minimizing ITZs interferences. This suggestion is probably not applicable to high performance concrete (HPC), because the density in the ITZ is exceeding that in bulk. In the case of HPC, the ITZ structure is largely improved due to the low range of w/c ratios (0.2~0.25) and addition of fine mineral admixtures.

Economic arguments demand volume fraction of aggregate to be as high as possible. For ordinary concrete, a value of 60% or so (for volume fraction) and a possibly wide grain size range (large value of d_m) are recommended for aggregate systems to strike a balance between economic considerations and concrete performance. At this aggregate density level, a relatively high tortuosity of the transport route can be guaranteed to outweigh the negative effects caused by interconnected porosities. Also, the surface spacing between aggregate grains will stay above the threshold value of ITZ interferences.

5. Conclusions

This paper studied by means of spacing parameters the spatial dispersion of aggregate grains (following Fuller size distribution function) in model concretes that were generated by the SPACE system. The simulation results indicate that the size range of aggregate grains has more pronounced influences on the spacing parameters than the volume fraction of aggregate in the investigated range of technical parameters. This is consistent with predictions of spacing parameters on the basis of stereological theory.

Mean free spacing $\overline{\lambda}$ is configuration-insensitive and therefore associated with structure-insensitive properties. In contrast, mean nearest neighbour distance Δ_3 and Δ_{3S}, is mainly governed by material configuration and can be applied therefore to structure-sensitive properties of concrete.

Aggregate systems should preferably be designed in the intermediate volume fraction range (up to 60%, or so) and with a wide size range. In this case, the interferences between ITZs of neighbouring aggregates can be maintained at an acceptable low level, avoiding significant negative influences on concrete permeability due to increased porosities connectivity. Otherwise ITZ should be upgraded by lowering water cement ratio and adding fine mineral admixtures.

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