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Stochastic heterogeneity as fundamental basis for the design and evaluation of experiments

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Abstract

This paper concerns the stochastic concept of heterogeneity of individual geometric descriptors of material properties. This allows defining the related concept of the representative volume/area element (RVE/RAE). A distinction is introduced between composition and configuration in geometric parameters, on the one hand, and structure-insensitivity and structure-sensitivity of material properties, on the other hand. Each configuration-sensitive geometric descriptor and each structure-sensitive material property will give rise to asymmetric probability density distributions for the relevant observations (on structure or property) obtained on equal-sized volume/area elements.

The paper discusses the biased character of data averaged for engineering purposes when based on sub-representative samples. Nevertheless, sub-representative elements can still be employed for comparative studies in concrete technology, however under the strict conditions that the sizes of the sample and of the smallest observed structural elements are maintained as fixed proportions of the corresponding RVEs/RAEs.

The implications of this underlying materials science concept are demonstrated in this paper by three illustrative examples dealing with cement-based materials.

Keywords: Composite material; Design of experiments; Heterogeneity; Representative volume and area elements; Size effect; Configuration- and structure-sensitivity

1. Introduction

Concrete is a particulate composite material on different levels of the microstructure. Gravel grains (and possibly macro-fibers) are aggregated on the meso-level in a cementitious matrix. Sand grains become discernable upon further increase of resolution in the aggregated mass of particles dispersed in the cement paste. An even more sensitive approach would allow detecting the very particles of this paste in the fresh state, or the hydrate structure of the hardened material. The molecular structure is situated at the lowest micro-structural level. The underlying concept of a *continuous range of micro-structural dimensions* as well as the three levels of aggregation, denoted by macro-, meso-, and micro-level in concrete technology, have been recognized for a long period of time in the physics and mechanics of deformable bodies [1,2].

Composite material behavior under forces reflects on the various aggregation levels the properties of composing parts of the material body and the material structure. This behavior is defined in terms of macro or engineering properties, such as the mechanical ones. Properties are denoted as structure-insensitive when solely governed by material *composition*, e.g. mass. On the contrary, structure-sensitive properties, such as crack initiation or brittle strength, are affected by the so-called group pattern or *configuration* of particles. As a consequence, particle size, shape and spacing are involved.

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Engineering properties can only be attributed to a material element of at least representative dimensions. Properties of sub-representative elements (i.e., smaller than the RVE/RAE) reveal an increasing degree of heterogeneity at reduced volume/area dimensions. This is reflected by the increasing width of the probability density curve of the descriptor of the material characteristic (structure, or property). However, each geometrical descriptor or parameter has its independent scale of heterogeneity due to different configuration sensitivity levels involved [1,2]. The same rule holds for properties with different degrees of structural-sensitivity, of course. Since RVE sizes of structure-sensitive properties can exceed those of structure-insensitive ones to a considerable degree, a wide range of RVE sizes will occur in practice [3,4].

So, heterogeneity is not a material characteristic, but it is a function of the descriptor selected to describe certain aspects of material structure, and of the linear size ratio of sample and RVE/RAE involved.

This latter could be denoted by sampling level or *sampling scale* (running from very small values to unity for representative elements).

2. Objectives and research significance

This paper discusses the fundamental notion of stochastic heterogeneity that governs observations on concrete structure and, as a consequence, also on concrete behavior as revealed in research on elements (three- or two-dimensional) of a particular size. Most important is the impact this has on *sampling strategy* and on *data evaluation*, both pursuing the generation of unbiased engineering information from material testing or quantitative image analysis approaches. Nevertheless, the common approach is sub-representative sampling in concrete technology, whereby obtained data will be biased unless sampling fulfils strict criteria discussed in this paper.

Three illustrative examples are additionally presented for the sole purpose of demonstrating the paramount importance of the stochastic heterogeneity concept for experimental design. Hence, this paper does not intend to discuss in depth the very topics covered by the examples. In the first example on *composition homogeneity*, focus will be on a recent *quantitative image analysis* approach to local porosity, a configuration-insensitive structural feature (volume fraction of pore space). The second example on *spacing homogeneity* will discuss an equally recent *computer-simulation* study of particle spacing, a material feature of configuration-sensitive nature. The third example deals with *brittle strength homogeneity*, a classical and well-known *analytical* approach to a weakest link material property (e.g., crack initiation strength, or brittle strength), thus being highly structure-sensitive. Material on the first two examples has been published earlier. The third subject is presented in its classical form, to demonstrate that the message of this paper has very old roots, indeed.

The stochastic concept of heterogeneity is well defined in material science textbooks; however, it generally neither underlies experimental design nor the evaluation of data in concrete technology. Engineering fracture mechanics in concrete technology recognizes this process of structural averaging (*globalization*) to yield biased information due to the so-called *size effect* (e.g., [5–8]). Yet, this is conceived an anomaly in engineering practice, because globalization is generally supposed to yield unbiased engineering information; but this is only guaranteed when based on representative samples.

The foregoing should have made it clear: a single size effect does not exist. Instead, size effects are linked up with anomalous outcomes of the globalization for engineering purposes of data obtained in sub-RVE (or, sub-RAE) sampling designs; a situation frequently occurring in concrete technology. The shortcomings of current practice are a source of inspiration for further research [9].

Experimental evaluation is commonly in terms of (sub-RVE) specimen size, and supposedly "homogeneous" material structure. Instead, it should be based on the different heterogeneity levels involved as a logical consequence of sampling the materials on specific aggregation levels of particulate matter [10]. This type of approach would *unify the engineering and materials science approaches*. In materials science approaches in concrete technology, e.g., pursuing the assessment of damage evolution characteristics under certain loading regimes, this heterogeneity (or size) effect is ignored in many cases, leading to misleading engineering information [11].

3. Fundamentals

3.1. Stochastic concept of heterogeneity

The value of a single descriptive parameter is determined on a series of similarly sized samples. These values are plotted as a probability density function. Heterogeneity is defined as the scatter (e.g., expressed by standard deviation) reflected by this probability density function of the selected parameter. Heterogeneity of a geometric parameter (such as crack length), or of the material property governed by this parameter, will increase for smaller samples [12]. But heterogeneity among *similarly sized volume elements* will also change as a result of differences in the observed micro-structural level of these samples (e.g., minimum crack length) implemented by differences in *microscope magnification* or *strain gauge length*. The involved ratio of minimum observed structural dimension and linear sample size could be denoted by *observation resolution or sensitivity*. To eliminate heterogeneity effects on sample averages, sampling scale as well as observation sensitivity should therefore have constant values in a comparative study. We will come back to this crucial evaluative statement in what follows. However, it shows already that insight into the size of the RVE/RAE is of utmost importance for the sampling strategy, and thus for the economy of the experiment. It is also essential for guaranteeing reliability of the experimental outcomes.

Globalization for engineering purposes of data derived from sub-representative samples yield *biased estimates* of structural features or of physico-mechanical properties, except for the special situation of a composition parameter or for the associated structure-insensitive property (i.e., density and mass, respectively). The literature provides for some materials outside the concrete field evidence for the dependence on material configuration of Young's modulus, though generally *assumed* structure-insensitive [13]. Additionally, however, differences have been detected for concrete with 16 mm maximum grain size between average values on meso-level (determined on a random set of 202 cubes of 20 mm size sawn from 250 mm cubes) and the corresponding value on engineering level (prisms with 200 mm cross section). Moreover, the probability density function for the mesoscopic values of Young's modulus was found markedly skewed to the left, reflecting its structure-sensitivity [4].

Biases will be more dramatic, of course, for some fracture properties and the underlying damage evolution characteristics, for which the dependence on structural details will be significantly larger. Fortunately, this high structure-sensitivity of the crack initiation strength will decline upon further energy dissipation due to micro-cracking, eliminating the high residual local stresses. Hence, the structure-sensitivity of the tensile strength is already reduced. As to the fracture energy, G_f, a situation of approximate structure-insensitivity will be attained, as has been proven by finite element approaches to SPACE-generated model concretes [14]. Also, a major part of the dissipated energy concerns the opening of the major crack of which the *tortuosity* is predominantly governed by volume fraction of the aggregate (with zero configuration-sensitivity) and not by the details of the sieve curve [15]. This supports the proposal as to the relatively small size of the RVE for G_f given in the final report of RILEM TC QFS [5].

Hence, representative, unbiased information on structure-sensitive (e.g., fracture-related) properties can be derived only from specimens of representative size for the very property at issue.

Otherwise, when working with sub-RVE samples, correlations should be established for all independent descriptive parameter observations with those of the associated RVEs.

In summary: heterogeneity in the geometric (structural) parameter that is considered relevant for the property of interest is reduced to an acceptable low level for the RAE. The RAE is therefore defined as *homogeneous for that particular structural (geometric) parameter*. The same can be stated for the heterogeneity in a property directly measured on samples of the material [1,4,16]. Thus, the degree of heterogeneity increases with a diminishing size ratio of sample and RVE/RAE [12].

A quite common misinterpretation of the heterogeneity phenomenon is revealed by a study performed by Keeton on cement paste, mortar and concrete [17]. Photo-elastic coatings of constant thickness that were glued on the surfaces of similarly-sized compressed specimens reflected for increasing grain size a growing disorder in shear strain contours under illumination by mono-chromatic light. Keeton erroneously associated this with heterogeneity as if dealing with a material property and classified the materials accordingly (e.g., heterogeneity in concrete exceeding that of mortar). Keeton's conclusion on increasing heterogeneity at larger grain sizes is incorrect, since strains were sampled by constant coating thickness, although the size of the RVE increased with grain size. So, at increasing maximum grain size, the size ratio of sample and RVE declined (i.e., sampling was achieved on lower sampling scale). This should inevitably lead to increased heterogeneity *as a reflection of the sampling strategy*. Hence, experiments only confirmed the validity of the theory. Heterogeneity should have been found similar when paste, mortar and concrete specimens had been observed at similar sampling scale (though, impossible by photo-elastic coating analysis).

3.2. Sensitivity to material structure

The shape of the probability density curve is symmetric (Gaussian) for a composition parameter or for a structure-insensitive property. However, the curve will be asymmetric (skew) in the case of configuration- or of structure-sensitivity [12]. Again, the width of the curve does not reflect the heterogeneity of the material, but merely of the scale of sampling. An experimental design based on constant sample (volume or area) sizes inevitably causes the *sampling scale to be different for different structural parameters or material properties*. For the same experimental design this would hold even for *similar* parameters or material properties (such as damage characteristics), but modified material structures, e.g., by increasing maximum grain size (as in Keeton's tests), or by prolonged hydration (porosity).

Fortunately, globalization of the data that are obtained on *sub*-RVE/RAE samples will not always yield biases in the engineering characteristics. Unbiased averages will be obtained in the cases of composition homogeneity, or structure-insensitive properties even when the scale of sampling would vary. However, this sampling strategy would be *improper* in the case of non-zero values of the degree of configuration- or structure-sensitivity. One is confronted with such conditions when investigating most fracture properties and/or the underlying geometric damage parameter(s). This is a highly relevant situation in materials research. Assessment of engineering properties on the basis of a sub-RVE sampling strategy requires therefore the availability of proper theoretical models, such as that of the weakest link on which Weibull's theory is based [18]. We will come back to this model in an illustrative example presented later. With a lacking theoretical basis, correlations should be established experimentally between sub-RVE sampling results and those obtained on relevant RVEs, conditions being similar, of course. And this has to be accomplished, basically, for each independent geometric parameter and associated material property, and for different environmental conditions.

4. Example I: composition homogeneity

Volume fraction of pores (attributed as porosity) in hydrated cement paste is chosen as a composition parameter (with zero configuration-sensitivity) to explore composition heterogeneity at different structural levels (i.e., due to different sampling scales) of cementitious materials. For more comprehensive information, see the relevant literature [12].

Porosity gradually declines during cement hydration since pore space is filled up with hydration products. Based on local porosity theory [19], Hu and Stroeven [12] studied in elements of ordinary cement paste with various water cement (w/c) ratios the changes during hydration in the distribution of local porosity. Section images of 263 × 186 μm were made by backscattered electron microscopy at optical resolution of 0.18 μm/pixel. These images were sub-divided into fields of constant size of which local porosity was determined, allowing construction of the probability density curve. The shape of this curve depends not only on technical parameters, but also on field size. Fig. 1 presents the local porosity distribution μ (=probability density curve) of a cement paste (w/c = 0.6) at different hydration times, maintaining according to common practice the same linear dimensions $L = 19$ μm of the square field. The differences between a relatively mature paste (at 14-day's hydration) and early-age pastes (at 3- and 7-day's hydration, respectively) are clear. The shape of the μ curves for pastes at 3 and 7 days of hydration are quite similar, with the curve for the 7-day's paste shifting to the left of the 3-day's curve. This shifting is due to the declining porosity as hydration proceeds. The shape of the μ curve for paste at 14-day's hydration indicates this paste to have the largest RAE (since this curve reveals the largest scatter/heterogeneity for the same L). Correspondingly, the heterogeneity of porosity in the cement paste is *seemingly* increasing during the hydration process, when based on fields of similar size.

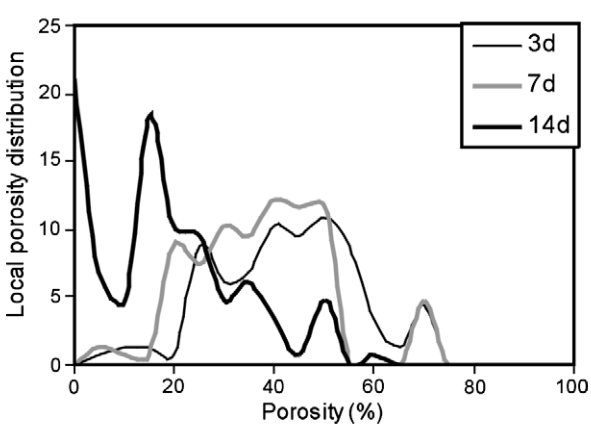

Fig. 1. Local porosity distribution (=probability density function) μ for cement paste with w/c = 0.6 at 3, 7 and 14 days of hydration, respectively, at constant linear field size $L = 19$ μm.

This conclusion drawn on evidence presented in Fig. 1 is incorrect (as in Keeton's case) because observations and comparisons were made on different structural levels. Cement hydration gradually changes the pore space from a connected structure to a de-percolated network with some isolated pores, yielding a significant decline in the number of pore features in the image plane. In the viewpoint of statistics, the smaller number of pore features leads to larger scatter of porosity data. This larger scatter is *seemingly* due to increased heterogeneity of porosity. In fact, the correct interpretation should be that it points toward hydration-driven increasing linear dimensions of the representative area elements (RAEs) for porosity.

Field sizes should be modified to the same proportion of the respective RAEs for a proper assessment of heterogeneity of porosity in cement pastes hydrated to different degrees. This issue of 'continuous scaling' is explicitly discussed in [10]. When the linear dimension of the RAE, L_{RAE}, exceeds the image size as in this study, the same 'level of microstructure' (i.e., the same coefficient of variation in the geometric parameter at issue) can be achieved by taking the ratio of field size L to L_{RAE} constant for all investigated specimens. These specific values of field size are denoted as L^* [12].

When the measurements are based on the same level of microstructure for the three cement pastes (at the appropriate values of L^*), the μ curves (normalized by their respective peak values) resemble each other far more closely than in Fig. 1 [12]. Theoretically, the three μ curves should be identical (curve with same scatter) and conform to the normal distribution (composition parameter) after eliminating the aging effect (by shifting the curves, so that the peak values of the three curves coincide).

The actual deviations of the experimental data from the normal distribution function are assessed by means of a χ^2-test [13,20,21]. For this purpose, the local porosity data are collected into traditional histograms. Details of the χ^2-test and the obtained results can be found in [12]. Fig. 2 represents the histograms of porosity distribution and the associated theoretical curves (normal distribution functions). The theoretical values of χ^2 exceed those of χ^2_{exp} for $n = 16$ and $n = 20$. Hence, at the 90% confidence level, we accept the hypothesis that in all three pastes porosities were distributed according to the same normal distribution curve. These observations are based on *images made with the same optical resolution*. If the observation sensitivity (resolution) had been also adjusted to the size of the RAE, the peakedness of the curves would have been increased with maturity, so that the curves in Fig. 2 would have matched even better. As a result, Fig. 2 confirms the concept of stochastic heterogeneity.

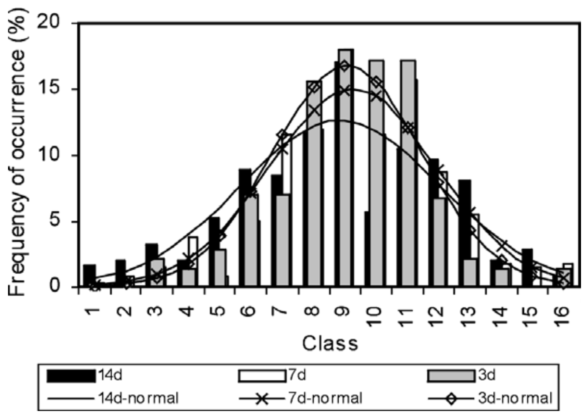

Fig. 2. Conventional frequency histograms of porosity for the same pastes as in Fig. 1 (with normalized mode values). However, in this case, field sizes L^* were proportional to the respective RAE sizes. The experimental values are indicated on basis of 16 porosity classes as columns and the approximated normal distribution curves are shown as continuous lines.

This example allows drawing the following conclusions:

(a) the probability density curves are Gaussian, confirming porosity to be structure-insensitive property, or when observed on images, a composition parameter;
(b) heterogeneity in porosity is the same in a quantitative image analysis approach to an hydrating system when the linear size ratio of sample and RAE (sampling scale) is kept constant, and
(c) when the observation/measuring sensitivity is adjusted to changes in the size of RAE.

5. Example II: spacing homogeneity

The second example deals with a particle spacing parameter of *moderately high degree of configuration sensitivity*, i.e., surface-to-surface spacing between nearest neighbors. This spacing parameter is denoted as Δ_{3s} in three-dimensional space, and Δ_{2s} in a section plane of the material body. Although the center-to-center nearest neighbor spacing, Δ_3, is defined in stereological literature [22], the surface-to-surface spacing may be more relevant for studying certain material properties. Δ_{3s} has been used earlier for studying the spatial dispersion of aggregate grains in concretes [23]. The surface-to-surface spacing may govern physical contributions in cementitious binder systems to strength, which have been demonstrated quite significant at lower water to cement ratios [24–26]. The surface-to-surface spacing of aggregate grains in combination with relevant information on extent and geometric features for a relevant configuration-sensitive spacing parameter of the interfacial transition zone (ITZ) can be used in modeling damage evolution in concrete [14,27]. An illustrative example will therefore be presented dealing with this highly relevant geometric feature. It will serve to demonstrate the effects of sampling strategy in a case of sensitivity to details in the material structure.

This issue is approached by computer simulation with the SPACE system that is based on a *concurrent algorithm* (particle interference is realized by a Newtonian system of colliding particles) [28,29]. Only such systems are able to properly simulate concrete microstructure allowing for "realistic" observations in the framework of structure-sensitive properties [30]. The conventional systems based on sequential particle addition algorithms [31–33] have been proven incapable of doing so [28,34]. Three aggregate structures of model concrete were generated with constant volume fraction of aggregate (50%). The aggregates conform in all cases to a continuous size distribution with a ratio of maximum to minimum grain size of two (i.e., diameters ranging from 5 to 10 mm). The linear dimensions of the reference frame (cubic container) were 253 mm, 92.6 mm and 46.3 mm, respectively, representing *different cases of sampling scale*. By using periodic boundary conditions for the container, the volume element will represent bulk material (no boundary disturbances). The dispersion of the aggregate grains in a mixture is characterized by the distribution probabilities of the spacing parameter Δ_{3s}, denoted as $f(\Delta_{3s})$. The surface-to-surface distances between all nearest neighbors were determined inside the reference frame and thereupon classified according to their length to obtain the probability density curve.

The probability density curves reveal differences with respect to "degree of heterogeneity" and "sample averages" as a function of the sampling scale. As to the latter aspect, Table 1 presents the mean and mode values of the spacing parameter Δ_{3s}. The experimental curves are approximated per specimen size by

$$y = axe^{-bx} \tag{1}$$

The values of a and b in Eq. (1) are determined by regression analysis. They are together with R^2 values (squared value of the correlation coefficient) presented in Table 2. The experimental distribution curves of the surface spacing and the fitting curves according to Eq. (1) are shown in Fig. 3 for the two extreme cases. Clearly, the probability density functions for Δ_{3s} are skew, with a concentration of relatively small values of the nearest neighbour surface-to-sur-

Table 1
Spacing parameters on different sampling levels (all dimensions in mm)

Container size	Mean spacing	Mode spacing
253	0.1083	0.0751
92.6	0.1062	0.0993
46.3	0.0989	0.1640

Table 2
Dimensionless regression parameters and χ^2-values (95% confidence level)

Container size (mm)	a	b	R^2	Scatter δ^2	χ^2-value
253	1.572	12.121	0.949	0.14	4.57
92.6	0.939	9.712	0.860	0.19	6.99
46.3	1.181	12.125	0.316	1.40	21.79

face distance. This is in agreement with theoretical predictions [35]. The scatter of the experimental values ($f_{i,\exp}$) around the regression curve is denoted as δ^2 and defined by

$$\delta^2 = \frac{1}{n-1}\sum_{i=1}^{n}(f_{\mathrm{i,exp}} - f_{\mathrm{i,reg}})^2 \qquad (2)$$

where $f_{i,\exp}$ and $f_{i,\mathrm{reg}}$ are experimental and regression values, respectively, of the interval probability (expressed in %) in class i of the histogram (number of classes is 50). Scatter (the degree of heterogeneity), δ^2, increases considerably at reduced sample sizes (Table 2; Fig. 3).

The sample with linear dimensions of 253 mm is considered as reference, supposedly representing configuration homogeneity of the aggregate grains. The differences between the reference histogram and histograms on sub-representative sampling level will incline at reducing sample size. The differences can be quantified by the aforementioned χ^2-value. The calculation results are presented also in Table 2.

The rule of thumb estimate for composition homogeneity is 4–5 times the largest structural dimension [36], i.e., 40–50 mm. Comparison of χ^2-values in Table 2 with a theoretical value of 3.325 (9 degrees of freedom at a confidence level of 95%), reveals the linear dimension of the RAE to be approximately 300 mm, exceeding to a considerable degree the estimate for composition homogeneity [20]. This is consistent with predictions of Brown [3] for composition and configuration homogeneity of conglomerates, and experimental observations on aggregate spacing by Stroeven [4].

This example allows drawing the following conclusions:

(a) the probability density functions $f(\Delta_{3s})$ are strongly skewed to the left. So, mean and mode values are different at different sampling scale levels (Table 1). Surface-to-surface spacing is a configuration-sensitive geometric parameter, indeed;
(b) serious biases result when mean or mode values are for engineering purposes derived from sub-representative sampling designs (Table 2); this is analogous to so-called 'size effects' in fracture mechanics testing;
(c) heterogeneity in the geometric parameter at issue (scatter in Table 2) increases considerably for the same material when sample size is reduced.

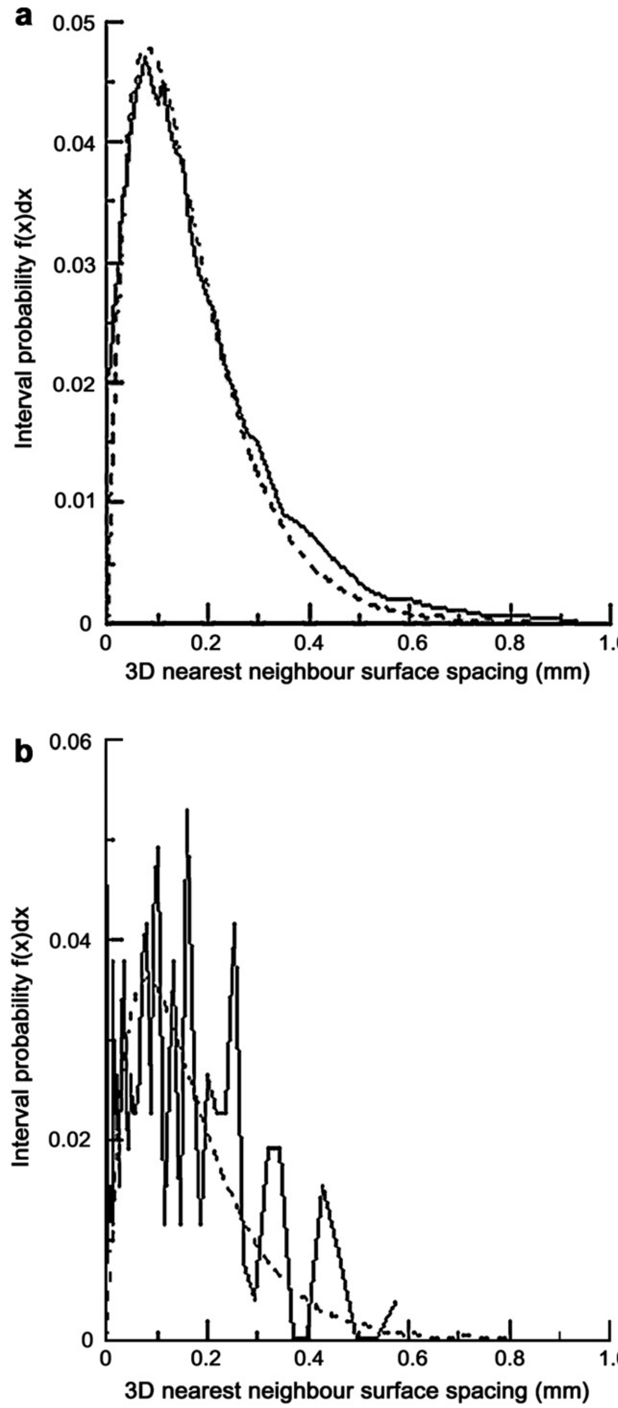

Fig. 3. Aggregate spacing in a single type of model concrete that was generated in elements of different linear dimensions, i.e., 253 mm (a) and 46.3 mm (b), respectively, is reflected by the experimental probability density curves of nearest neighbor surface-to-surface spacing, $f(\Delta_{3s})$ (solid lines), and corresponding regression curves (dashed lines).

6. Example III: brittle strength homogeneity

The interest in the stochastic background of weakest link problems goes back about one century [37], and major statistical contributions to estimating fracture properties, recognizing their structure-sensitive background, appeared during the 1920s to 1940s. Analyzing the opinions at that time, Epstein [38] concluded "Many scientists accept the fact that the Gaussian distribution plays a fundamental role in science and, in fact, there are many who feel that this is the only distribution which nature calls truly her own... however, in a certain class of phenomena the characteristic distributions are far from normal and are, in fact, strongly skewed to the left". Although this seems more widely accepted nowadays in engineering approaches to fracture problems, the underlying heterogeneity concept is still widely ignored, and the implications missed. That is the very reason to take an example with such old roots; what is presented is therefore far from new, but by many somehow overlooked or at least underestimated as to its consequences. Therefore, the example will also be presented in its "classical" form.

It is commonly assumed in the case of concrete that flaws in the material body are at the basis of fracture properties. For the present illustrative purpose, the crack initiation (or brittle) strength is selected as a property highly sensitive to material structure. Nowadays, flaws are associated in normal concrete with more porous areas in the ITZ around aggregate particles. Weibull [18] has elaborated a theoretical concept for brittle fracture based on the weakest link concept for elementary units containing single flaws.

The probability density function of the brittle strength of sub-volumes of the material body containing a single elementary flaw (here referred to as elementary units) is denoted by $f(\sigma)$, and the cumulative frequency curve by

$$F(\sigma) = \int_{-\infty}^{\sigma} f(k)\mathrm{d}k \tag{3}$$

When the material body encompasses a total number of i elementary units, the probability density function $g_i(\sigma)$ of the *minimum value* of brittle strength is given by [39,40]

$$g_i(\sigma) = i \cdot f(\sigma)[1 - F(\sigma)]^{i-1} \tag{4}$$

with the cumulative distribution function $G_i(\sigma)$

$$G_i(\sigma) = \int_{-\infty}^{\sigma} g_i(k)\mathrm{d}k = 1 - [1 - F(\sigma)]^i \tag{5}$$

Determination of the median value of brittle strength $\tilde{\sigma}$ is straightforward [37]. The mode $\hat{\sigma}$ is obtained upon differentiation of Eq. (5) and equating to zero. The resulting equation cannot be solved in an elementary way for *normally distributed brittle strength values* of the elementary units. A new variable z is therefore introduced and defined by $z = i \cdot F(\sigma)$. The cumulative and probability density functions of z can be obtained for the limiting case of $i \to \infty$

$$G_{i\to\infty}(z) = 1 - \mathrm{e}^{-z} \text{ and } g_{i\to\infty}(z) = \mathrm{e}^{-z} \tag{6}$$

An asymptotic development allows expressing σ into z [41–43]

$$\sigma = m - s\left[\sqrt{2\ln i} - \frac{\ln\ln i + \ln 4\pi}{2\sqrt{2\ln i}} - \frac{\ln z}{\sqrt{2\ln i}}\right] \tag{7}$$

in which m and s are the mean and standard deviation of the brittle strength of the elementary units, governed by the normal distribution function $f(\sigma)$.The most probable value (mode) and variance of σ are given respectively by

$$E(\sigma) = m - s\left[\sqrt{2\ln i} - \frac{\ln\ln i + \ln 4\pi}{2\sqrt{2\ln i}}\right]$$

$$\text{and } D^2(\sigma) = \frac{\pi^2 s^2}{12\ln i} \tag{8}$$

The mean and standard deviation of the material containing a very large number of flaws can be formulated by $\hat{\sigma} = m - \alpha_i s$ and $s_i = \beta_i s$ in which the coefficients α_i and β_i are given by

$$\alpha_i = \sqrt{2\ln i} - \frac{\ln\ln i + \ln 4\pi}{2\sqrt{2\ln i}} \text{ and } \beta_i = \frac{\pi}{2\sqrt{3\ln i}} \tag{9}$$

Tables are available in the literature for α_i and β_i [44]. The decline rate in scatter with increasing sample volume is exceeding the one in strength. So, also the coefficient of variation is declining with increasing sample volume. In other words, sampling sub-volumes of the RVE (defined at an acceptable level of scatter) will lead to greater strength values accompanied by *disproportionately* increased scatter (or, *heterogeneity*).

The strength ratio of an arbitrary material volume and the RVE is obtained by means of Eqs. (8) and (9), yielding

$$\frac{\bar{\sigma}}{\bar{\sigma}_{\mathrm{RVE}}} = \frac{m - \alpha_i s}{m - \alpha_{\mathrm{RVE}} s} \approx 1 - \frac{s}{m}(\alpha_i - \alpha_{\mathrm{RVE}}) \tag{10}$$

with

$$\alpha_i - \alpha_{\mathrm{RVE}} = \left(\frac{\beta_{\mathrm{RVE}}}{\beta_i} - 1\right)\alpha_{\mathrm{RVE}} \tag{11}$$

Hence, the strength increase due to sub-RVE sampling strategy is reflected by

$$\frac{\bar{\sigma}}{\sigma_{\mathrm{RVE}}} \approx 1 + \frac{s}{m}\left(1 - \frac{\beta_{\mathrm{RVE}}}{\beta_i}\right)\alpha_{\mathrm{RVE}} \tag{12}$$

Eq. (12) depicts the *strength increase by stochastic heterogeneity* on sub-RVE sampling level. The value of β_{RVE} indicates the acceptable scatter limit ($=s_i/s$) for declaring the sample volume representative for brittle strength, and thereby defining it as homogeneous. Heterogeneity is expressed by the ratio $\beta_i/\beta_{\mathrm{RVE}} = s_i/s_{\mathrm{RVE}}$. The microscopic material parameter $(s/m)^2 = v$ has been referred to in the international literature as the unit coefficient of variation [45]. When β_{RVE} is selected, α_{RVE} is given by the relationship $\alpha_{\mathrm{RVE}} = \pi/(\sqrt{6}\beta_{\mathrm{RVE}})$.

Upon combination of Eqs. (9) and (12), the ratio of linear dimension of sample and the RVE (proportional to $\sqrt[3]{i/i_{\mathrm{RVE}}}$) is introduced as the running parameter. Hence

$$\frac{\bar{\sigma}}{\sigma_{\mathrm{RVE}}} \approx 1 + \frac{s}{m}\alpha_{\mathrm{RVE}}\left(1 - \frac{\sqrt[3]{\ln i}}{\sqrt[3]{\ln i_{\mathrm{RVE}}}}\right) \tag{13}$$

This defines the so-called *size effect* [5], in the present case supposedly for the crack initiation strength (Fig. 4). The unit coefficient of variation in Eqs. (12) and (13) is available for adjusting to experimental data. Note again that this paper is not intended, however, to discuss statistical strength theories.

Of course, qualitatively similar conclusions can be drawn as for the second example

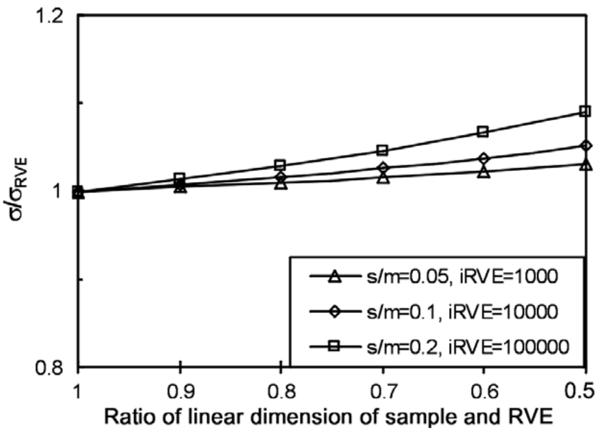

Fig. 4. Overestimate of brittle strength on sub-RVE sampling level. i_{RVE} stands for the number of flaws in the RVE. m and s are the mean and standard deviation of the normally distributed brittle strength values of the elementary units containing single flaws.

(a) the probability density function of $\hat{\sigma}$ is strongly skewed to the left (Eq. (9)); we deal with a performance parameter that is extremely structure-sensitive. Moreover, ample evidences can be found in the literature on the skewness of the probability density function of concrete's tensile strength;
(b) serious biases in engineering strength parameters (mode, scatter) result from sub-RVE sampling designs (Fig. 4); these are the so-called 'size effects';
(c) heterogeneity in the strength parameter (Eq. (9)) increases disproportionately at reduced sample size, so also "coarseness" (coefficient of variation) increases;
(d) heterogeneity in the strength parameter (Eq. (9)) is also influenced by the size of the smallest crack incorporated in the study (influencing i in Eq. (9)), so by the observation sensitivity or magnification. This is also reflected by Fig. 4.

7. Summary and conclusions

Heterogeneity is not a material characteristic. Instead, it is a stochastic concept that governs the design of experiments; it inevitably results from sub-representative sampling procedures, either in 3D (material testing) or in 2D (image analysis) approaches. Homogeneity of a geometric parameter or of the associated material property can only be achieved for elements that are large enough to reduce the between-samples scatter in the relevant parameter or property to an acceptable level. This is the so-called representative volume/area element (RVE/RAE). When the experimental design is based on volume (in engineering testing) or on area elements (in quantitative image analysis) of sub-representative size *as to the property or geometric feature of interest*, the associated degree of heterogeneity is a function of the size ratio of sample and representative element (sampling scale).

Hence, biases will be similar in *comparative studies* on cementitious materials executed under equal conditions when this size ratio is maintained at constant level. This implies that sample size has to be adjusted to changes in the size of RVE or RAE due to material modifications (e.g., due to different maximum grain sizes in concrete, or to different maturities of cement paste). So, the requirement of constant size ratio of sample and RVE/RAE (sampling scale) will generally require experimental designs encompassing different sample sizes. Globalized data on heterogeneous parameters (for engineering purposes) obtained in sub-representative sampling designs are inevitably biased; this is leading to a parameter-related "size effect", as is gradually being recognized in fracture mechanics testing. In engineering testing (employing volume elements), as well as in materials investigations (based on area elements), all results should be corrected therefore to obtain unbiased engineering estimates. These correlations should be either obtained experimentally or analytically, based on models of the material in which heterogeneity is a continuous function of the size ratio of sample and RVA/RAE.

Geometric parameters can reflect to a certain degree material configuration. When this dependence is zero, the parameter is defined as a composition parameter (e.g., volume fraction). In other cases, we deal with a configuration-sensitive parameter that will depend on the group pattern of relevant particles (size, shape, dispersion). The same holds for the material property associated with certain features of material structure. These properties range between structure-insensitive (e.g., mass) and structure-sensitive (e.g., crack initiation strength). The size of the RVE/RAE is a direct reflection of the degree of sensitivity to configuration of the geometric parameter, or to structure of the material property. This implies each geometric parameter and each material property *to have its own independent scale of heterogeneity* and its own – by definition – homogeneous RVE/RAE with dimensions specific for that parameter/property.

Heterogeneity effects on global estimates in *comparative studies in materials testing* as well as in *comparative studies by quantitative image analysis approaches*, can be excluded by maintaining the size of the sample and of the smallest structural elements (grains, pores, cracks) as constant fractions of the linear dimensions of the RVEs/RAEs involved. Only under such conditions can the experiments be analyzed in an unbiased way. That would constitute the reliable and economic fundament for engineering approaches pursuing technological progress.

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