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Stereological Approaches to Cementitious Composites

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A brief survey, based on illustrative examples, will be presented of the various types of structural problems, that have been solved by applying statistico-geometrical methods in this field. Experiments will take up the major part of it. Petrographic, metallographic as well as quantitative macroscopical applications will be treated. They deal with all possible components of the (wire-reinforced) cementitious composites. Form is not playing a major role, however. A single, theoretical application will be mentioned to formulating the solution of the particle interlock problem. A more recent tool - still under development - is the computer-simulation of structures of (cementitious) composites. Sofar, we have studied particulate systems with spherical aggregate. However, we aim at introducing differently shaped particles.

INTRODUCTION

Concrete is a macroscopically heterogeneous, brittle material. This has major implications for sampling and data acquisition. The representative volume or area element has macroscopical dimensions. Extreme care should be bestowed on specimen preparation in order not to disturb the structural features (basically, pressure, temperature changes and water produce such effects, while alone or in combination indispensable for the preparation). Mostly data acquisition can only be realized when optical contrast in enhanced. Further, the concrete structure is formed in situ. Prefabricated or not, this leads to a relatively large structural variability.

Various techniques for quantitative image analysis are in use in concrete technology. They can be distinguished in petrographical, microscopical and macroscopical methods. In an illustrative way they will successively be treated. Stereology also renders possible describing statistico-geometrical problems met in concrete technology. A single example will be given.

CONCRETE PETROGRAPHY

A first logical step towards the establishment of a structural basis for physico-mechanical behaviour is concrete petrography. Petrographic examination of hardened mortar or concrete - a man-made rock - aims at describing by the descriptive methods of petrography the formation and composition of the mortar or concre

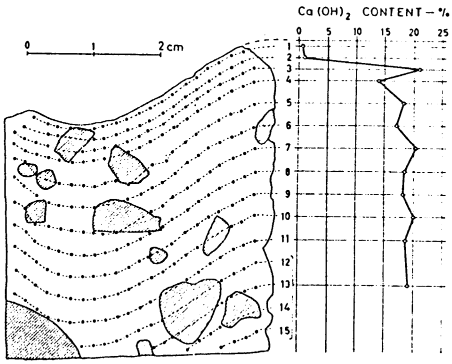


Fig. 1. The amount of $Ca(OH)_2$ in concrete analysed by quantitative petrographic means (Larsen).

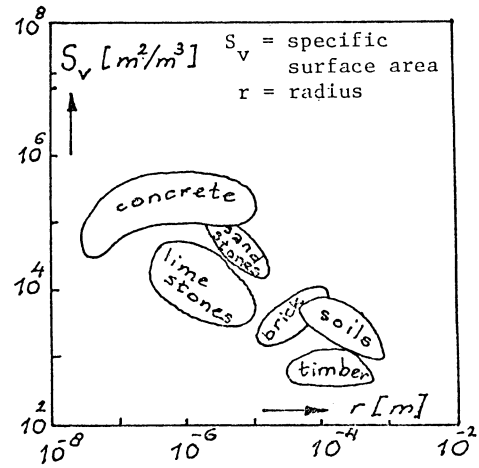


Fig. 2. Pore properties of commom construction materials (Hayes).

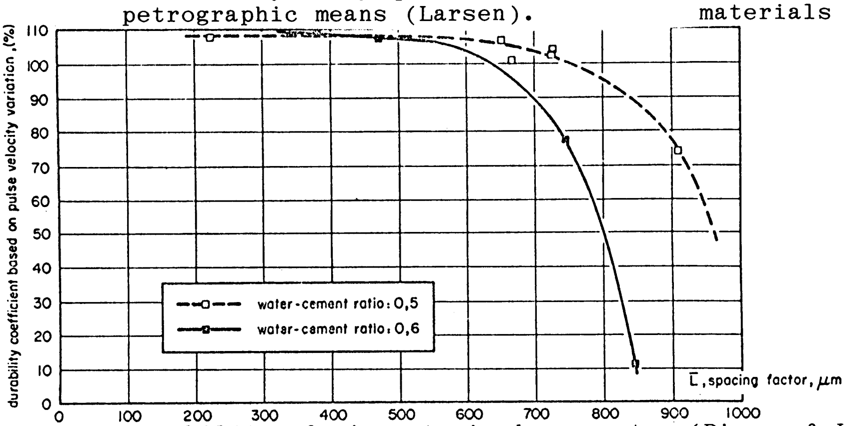


Fig. 3. Durability of air entrained concretes (Pigeon & Lachance).

te and to classify it as to type, condition, and serviceability. Such microscopical observations date back to the beginning of the century as far as the aggregate is concerned. Recommended practices for petrographic examination are available. The use of the microscope is even older. One century ago Le Chatelier published what is considered a first example of such an approach. A major problem treated by concrete petrographers is that of the alkali-aggregate reaction. the petrographic approach has been standardized in ASTM 295-79. Particular products in concrete are reactive in the alkaline environment of pore water solution. Sole detection of these products mostly suffices. An example of quantitative image analysis in concrete technology is the study of the amount of $Ca(OH)_2$ by Larsen. In a concrete dam in Sweden percolation of water along fractures was observed. Analysis of water seeping through showed that $Ca(OH)_2$ was leached from the concrete along the cracks.

A direct reduction in matrix strength can be the result of this disintegration. $Ca(OH)_2$ also preserves the alkalinity of the matrix so that leaching can cause the carbonatation process to be accelerated, finally leaving the reinforcement unprotected. Fig. 1

presents a section image and measuring system used in the study. Point counting (or point "measuring" as the author calls it, since line elements of the scale are counted) is performed by shifting the scale of the eye piece in such a way that only the cement paste is covered. In doing so, the amount of $Ca(OH)_2$ in the cement paste is determined. Since $Ca(OH)_2$ tends to concentrate in the curved interphase area of particles and matrix - an area that is less accessible for measuring - the data will be systematically on the low side. This will not have influenced the outcomes, however, that revealed leaching to be concentrated in a narrow area bordering the cracks. Since most of the investigations can be of a qualitative nature, in petrography only seldom use is made of quantitative image analysis techniques.

METALLOGRAPHIC APPROACHES

Metallographic (quantitative microscopical) methods were first introduced to evaluate the pore structure in concrete around the end of World War II. Areal, lineal and point count techniques have been applied for that purpose. As the practice of entrained air in concrete to enhance the freeze-thaw and de-icing salts resistance has become more wide-spread, there has sprang up a drastically increased interest in means to determine the air content in concrete (mortar, brick work, tiles, etc.). We may refer here as an example to the life-long activities in the field by the Mather's. A symposium will be dedicated in 1987 to their contributions to durability of concrete encompassing petrographical and microscopical investigations.

Durability of porous building materials is particularly governed by their porosity. The determination of pore properties is therefore a vital step in the evaluation of material and in quality control of manufactured products. It is of scientific as well as of economic significance. The three structural properties fundamental to the description of porous building materials are pore content, specific surface area, and size distribution. The wide spectrum of these pore properties encountered in practice in a series of common building materials is illustrated in fig. 2. A survey of the available techniques of pore analysis is presented by Hayes. Among other things, it is shown that stereological techniques take up only a modest position.

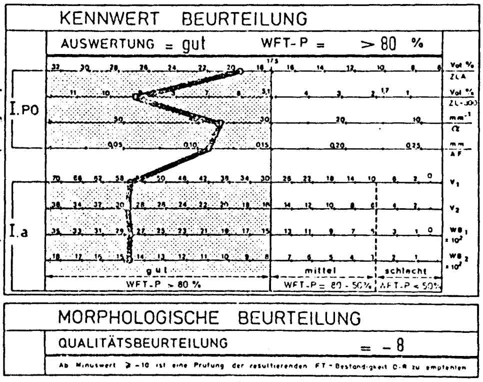
ASTM C 457-80 gives a prescription for the "microscopical determination of air-void content and parameters of air-void systems in hardened concrete". The linear traverse (Rosiwal) and modified point-count methods are employed for that purpose. Of paramount importance is this approach for predicting the frost-thaw-de-icing salts resistance of porous building materials. Entrained pores are almost spherical with diameters between 2 en 10 μm and a number of millimeters. The pores smaller than 0.3 mm are of particular relevance. Size and shape of this type of pores make the use of stereological techniques for quantitative image analysis very appropriate.

The pioneering work by Powers still constitutes the basis for the determination of the durability of porous materials. He proposed a critical spacing factor that is incorporated in the ASTM prescription. This factor $\bar{L}$ is, however, based on a monosized cubic lattice of spherical pores. The mechanism of water transport through capilary pores towards the macro pores would suggest the average nearest neighbour distance to be a very suitable spacing factor that is based on a more realistic pore distribution. Nevertheless, Powers' spacing factor is also proportional to interpore spacing. As an example, fig. 3 presents results taken from a recent publication, particularly emphasizing the effect of the freezing rate on durability. It is shown that the normally adopted critical spacing factor of 0.25 mm is increased when the freezing

Texture components		Defects Classification	Index	Remarks
Macro-porosity *)	Agglomer. of voids	medium	-2	
	Compaction voids			
	«Water voids»			
Cement paste capillarity	Uniform	subjective	-1	Medium
	Variable	subjective		
Cracks in cement paste *)	≥ 0.005 mm wide	few	-1	Larger,
	< 0.005 mm wide	medium	-1	Cracks
Aggregates **)	Cracks and Porosity	medium	-2	
Bond between aggregate and cement paste **)	Capillarity defects			
	Bond def. due to cr.	few	-1	
	Agglom. of voids Compaction voids «Water voids»			
Hydration		subjective	0	Good
Quality assessment		Total	-8	If negative results ≥ -10 a frost resp. a frost-de-icing salt test D-R is advisable.

Evaluation of the number of defects according to the analytical criteria:

Defects, classification		Defects, maximum number/cm² *) In liant Z		**) In the aggregates (100-Z)	
Few	≤	Z × 0.4 =	13	(100-Z) × 0.133 =	9
Medium	≤	Z × 2.0 =	63	(100-Z) × 0.666 =	46
Frequent	>	Z × 2.0 =	63	(100-Z) × 0.666 =	46
Z = hardened liant		C + W + LA in vol. %		=	31.3



ZLA=air content hardened concrete
ZL-300=volume of pores, r ≤ 300 μm
α =specific surface area pores
AF=spacing factor

Fig. 4. (left) and 5. (right)
Qualitative and quantitative microscopy at LPM.

rate is reduced. Modern usage of very fine powders in concrete, such as silica fume, has demontrated that the spacing factor alone is unsuitable for predicting durability properties of such composites.

Centers dealing with this type of durability problems are mainly located in the Nordic countries (Scandinavia, Soviet Union) and Japan, Canada and the USA. Mountainous areas suffer from the same problem. Hence, a lot of work has been performed in European countries like West Germany, Switzerland and Austria. In Scandinavia, the "Technologisk Institut" (TI) in Tastrup, Denmark takes up a leading position. In Central Europe, the "Labor für Preparation und Methodik (LPM) in Beinwil am See, Switzerland is to be mentioned. Both institutes offer full support to the building industry, in particular to the road and airport construction industry. Both institutes are equiped with modern automatic analysers (Leitz's TAS).

The LPM employs the so-called Romer-Dabrolubov method. The microscopic investigations of concrete are carried out on thin sections (20-30 μm, 800 mm^2) made from concrete cores (50 mm in diameter) impregnated with a resin containing a special fluorescent dye and examined under the microscope in transmitted ultraviolet light. In this way the three concrete texture components (aggregates, hardened cement paste and voids (20 μm)) can easily be distinguished. Qualitative morphological investigations are also executed on the same images. Both data sets are used for quality control purposes (figs. 4 and 5). A similar approach is followed for other cement-based materials, ceramics, bitumes/-asphalts, rock material, sand stone, etc.

Specimen preparation is a tedious job. Walker & Marshall claim to need four days for the preparation of large-size ultra thin sections of 5 μm. By processing a number of them time investment could be brought back to 1.3 man-hours. Willis, Dobrolubov and Romer need five hours for "preparation and a comprehensive pore analysis with a concrete morphological determination" and six to eight hours for an analysis by the linear traverse method. A condensed survey of all problems met with in specimen preparation and their solution is presented by Kleine-Bünger & Scheer of the "Staatliches Materialprüfungsamt" in Dortmund.

The TI applies polished sections as is also prescribed by ASTM C 457. In the latter case, however, no prescription is presented for enhancing the structural contrast. In Tastrup this technique is improved by stamping the specimen surface with a black stamping fluid, whereupon the pores are filled with ZnO + vaseline. Finally, a thin layer of powdered gypsum is applied to cover the pore filling. The result is a high contrast pattern (fig. 6). Chatterji & Gudmundsson claim that approximately two hours are required for the complete procedure of grinding, specimen preparation and image analysis. Of course, this approach is exclusively tuned to pore analysis. The ASTM procedure has been improved in various institutes by impregnation with coloured resins. In the "Forschungsinstitut der Zementindustrie" in Düsseldorf, Schäfer and Walz & Derfert used a two-component epoxy containing a dispersed red-coloured pigment. Finally, a foil-print technique has to be mentioned. Flat polished surfaces of specimens (without pore or crack filler) of concretes or bricks can directly be copied to yield good contrast patters (Blümel).

QUANTITATIVE MACROSCOPY

The approaches collected under this heading deal with dispersion problems of second-phase elements in the cementitious matrix. Possible elements are: particles, wires and cracks. The same stereological techniques as applied in quantitative microscopy or metallurgy can be (and sometimes are) employed on macro level. Although early investigations of the defect structure in concrete go back as fas as 1927, the pioneering work in the field undertaken at Cornell University appeared in the early thirties. Examples of quantitative evaluations are quite scarce. The present author has contributed to this field. A further theoretical contribution is due to Oda.

Concrete always contains a vast amount of micro cracks. The number of cracks in virgin specimens may amount to 50% or more of those present at rupture. An increase in total crack length is mostly associated with a reduction in residual strength. Objective means for crack morphometry are therefore required. Suitable methods for contrast improvement come from petrography. Commomly, coloured or fluorescent inks are used for that purpose. Unfortunately, in most investigations crack patterns are only analysed in a qualitative way. An example of a semi-quantitative image analysis is presented by Gjorv & Shah. The authors count the number of intersections with a superimposed test line. The specimens were previously subjected to durability tests. Crack were visualized by red ink penetration. Applying stereological theory, they could have determined the specific surface area from their results.

The present author has extensively applied stereological techniques in this field. Visualization was achieved by the mentioned filtered particle method (fig. 7). Slides were prepared and thereupon projected on a semi-transparent screen for manually counting of intersections with a superimposed line grid. Further, the number of cracks was counted. A good example of fruitful use of this approach can be found in Stroeven (1978). Herein, tests are described in which the low-cycle fatigue behaviour of concrete (important for off-shore structures) was studied. The acting crack

Fig. 6 Surface of concrete specimen revealing white pores filled with ZnO + vaseline and gypsum (T.I.).

Fig. 7 Cracking in concrete visualized by filtered particle methode (Stroeven).

mechanisms under different load amplitudes were analysed by means of stereological techniques. Large differences were found in the roses of the number of intersections, offering the solution to the posed problem, since changes in other morphologic parameters were less striking.

New types of concrete are being brought onto the market, among other things fibre reinforced concrete (mortar). Fibres improve strength and, particularly, toughness. Due to fabrication and densification techniques the fibre structure reveals segregation and partial orientation. The present author developed the framework for reconstructing the 3-D features of steel fibre reinforced concrete (SFRC) from measurements in the section or projection (X-ray radiography) plane. A combination of independent observations can suit the purpose. Feature counts or intersection counts in the projection plane form the stereological basis.
Quite independently, Kasperkiewicz has developed geometrical-statistical formulas to define fibre spacing in particular for thin elements. For similar cases results are identical.

On the same macroscopical level other outgrowths of metallographic techniques can be classified. The objectives are to analyse the distribution of particles in space. To that end, this author used mono-sized ceramic spheres. Areal, lineal and random secants analyses were executed. Similar problems were faced by Pigeon. He used glass marble aggregate for this purpose, however. Moreover, unaware of stereology he experimentally derived a relationship between areal and volume fractions. Gast considered the problem of particle dispersion in concrete analogously to pore size distribution problems. Wu & Karl studied the effect of vibration techniques on grain inhomogeneity and anisometry. Sections of cubes were to that end examined. The effect of the vibration technique was convincingly demonstrated. Quantitative analysis incorrectly assumed the particle size distribution to be directly governed by the observed section size distribution. Anisometry was determined by measuring the principal directions of the grain sections! Saltikov's method of directed secants could have proved its value.

The derivation of constitutive relationships, in which structural information is incorporated, requires describing the statistico-geometrical characteristics of the concrete composite. An example of such an approach is the so-called particale interlock problem in concrete technology. Walraven approached the problem correctly. Unaware of stereological theory, however, he solved the well-known integral equation of Abel's type by numerical means. This author demonstrated that analytical solutions can be derived for this problem. Moreover, the two-dimensional solution by Walraven can easily be obtained by assuming the particles to be squares in a plane, as demonstrated by this author. Finally, it is worth mentioning computer-simulation as a powerful tool in structural problem solving. Preliminary results were obtained by this author, demonstrating the potentiallity of the very method. Further development is pursued by considering differently shaped objects.

CONCLUSIONS

Stereological techniques can fruitfully be used to quantitatively analyse structural problems met with in concrete technology on micro as well as on macro level. The various approaches are des-

cribed by presenting some relevant examples.
Stereological theory can also be employed for the derivation of constitutive relationships in which structural information should be incorporated.

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