The paper presents a method of pore connectivity analysis applied to specimens of cement based composites differing in water to cement ratio. The method employed X-ray microtomography (micro-CT). Microtomography supplied digitized three-dimensional radiographs of small concrete specimens. The data derived from the radiographs were applied as an input into the application based on the algorithm called ‘random walk simulation’. As the result a parameter called diffusive tortuosity was established and compared with estimated porosity of examined specimens.

Keywords: X-ray tomography, concrete microstructure, diffusive tortuosity

1. Introduction

The research on durability of concrete outdoor structures is closely related to their properties of transport of potentially harmful ionic fluids and aggressive gases. The properties mentioned above strongly depend on the morphology of the pores inside the described material. In last years the micro-CT method was widely applied to get 3D digitized data sets useful to study a variety of issues related with concrete microstructure and its evolution due to ageing processes [1,2]. Fig. 1 presents examples of microstructure obtained with the application of micro-CT method of two brittle matrix composites extremely differing with their property of pore interconnectivity. The 3D images present a cubes of 3×3×3 mm reconstructed with the resolution of 5 μm per voxel (volumetric pixel).

Several authors applied the micro-CT technique in their research of concrete. The authors of present paper compared the average mortar density and volumetric pore distribution of three different concretes in [2] (2014). Garboczi [3] (2002) applied the micro-CT technique to reconstruct the aggregate shapes. He investigated concrete 108×108×108 mm cubes with voxel resolution of 0.4 mm³. The data obtained from the scans of the concrete samples enabled the author to approximate the real shapes of the recognized aggregates by its mathematical models to built the stereological model of entire population of aggregates. Lanzón et al. [4] (2012) used the
micro-CT method to test the specimens made with crushed limestone aggregate (82.80 wt.%), cement CEM II/B-L 42.5R (16.50 wt.%) and additions (0.60 wt.%). Three low-density additions significantly different in parameters of their granulometric features have been used, i.e. their medians and variation coefficients were different. The low-density additions were: expanded perlite, expanded glass and cenospheres (hollow microspheres). The scanning was performed on cylinders of 8 mm in diameter and of 8 mm of height cut off the bulk of the mortars. The resolution of 8 $\mu$m per voxel was achieved in resulting tomograms. The authors have determined capillary water absorption for mortars with the three different low-density additions and for the reference mortar by partial immersion of the samples in water. The results agreed with the volumetric pore concentration data obtained by processing of test results using micro-CT. The correlation coefficient of 0.964 was obtained between the air pores content determined using micro-CT and the capillary water absorption. That nature of the mortars allows for precise segmentation of air pores since air is clearly differentiated from the solid and gelled components. Stock et al. [5] (2002) analysed the progression of sulfate attack within the cement paste. In the paper they presented the microstructure of their specimens. The specimens were the cylinders of 10 mm diameter and of 40 mm length. The resolution of rendered images was 20 $\mu$m in cross-sections and 30 $\mu$m in thickness. The recognized damage levels in specimens exposed to a $\text{Na}_2\text{SO}_4$ solution in different conditions varied from 0 (no damage) to 4 (extreme damage). Nakashima and Kamia [6] (2012) derived transport coefficients were different. The low-density additions were: expansion perlite, expanded glass and cenospheres (hollow microspheres). The process of digitizing of the concrete specimens reconstructed every 5 $\mu$m of specimen height, 2000$\times$2000 of pixels each as a result of scanning of a $9 \times 9$ mm and $H = 9$ mm cylinder with a resolution of 5 $\mu$m per voxel. With the coding scheme of 1 byte per pixel, the total number of bytes in such a dataset would be 7,200,000,000 bytes. It is inconvenient to be processed in one piece in computer memory. To make the image processing possible the usual procedure used by the majority of the authors cited, is to extract a subset of data, preferably of cubic shape for further computations. The resulting subset is called a ROI (Region Of Interest) and is immune from the cracked areas situated in proximity of the specimen surface usually damaged by a drilling tool. The authors of present paper have obtained 1800 crossections of the data obtained from several specimens into one set of data. The limitation of dimensions is the argument to limit to the matrix without coarse aggregate grains.

2. The algorithm of determining of difussive tortuosity

The process of digitizing of the concrete specimens results in binary datasets of consecutive crosssections. The authors of present paper have obtained 1800 crossections (reconstructed every 5 $\mu$m of specimen height), 2000$\times$2000 of pixels each as a result of scanning of a $9 \times 9$ mm and $H = 9$ mm cylinder with a resolution of 5 $\mu$m per voxel. With the coding scheme of 1 byte per pixel, the total number of bytes in such a dataset would be 7,200,000,000 bytes. It is inconvenient to be processed in one piece in computer memory. To make the image processing possible the usual procedure used by the majority of the authors cited, is to extract a subset of data, preferably of cubic shape for further computations. The resulting subset is called a ROI (Region Of Interest) and is immune from the cracked areas situated in proximity of the specimen surface usually damaged by a drilling tool. The authors of [7] analysed ROIs containing 255$^3$ voxels. In [8] 1000$^3$ voxels were processed. The authors of present paper decided to process 6 different ROIs of 216,000,000 voxels each. That way the ROIs described in next paragraphs represented cubes of $3\times3\times3$ mm. To analyse the internal pore network connectivity a random walk algorithm [7] was implemented in a form of originally prepared executable program for the PC computer. The latter algorithm originally was intended to simulate the diffusion of gases and liquids in the interconnected network of pores. At the starting point of the...
action a certain number of ‘walkers’ was distributed randomly across the space reconstructed by means of the micro-CT scanning procedure. The ‘walkers’ occupy one voxel of space and can be understood as a marked point in the dataset representing the analysed volume of the specimen. The walkers migrated on neighbouring voxels obeying the information on voxels brightness (i.e. material density). The walkers executed jumps in randomly chosen direction but the jumps could be performed if the neighbouring voxel belonged to a pore and otherwise the jumps were discarded. After refreshing the position of all walkers, one epoch of its action was completed by the algorithm. The number of epochs was measured by the dimensionless integer time \( t \). The primary output of the random walk procedure is the walkers mean-square displacement \( < r(t)^2 > \) as a function of time \((x_i, y_i, z_i)\) are the coordinates of a current walker position and \( n \) is a number of operating walkers:

\[
<r(t)^2> = \frac{1}{n} \sum_{i=1}^{n} [(x_i(t) - x_i(0))^2 + (y_i(t) - y_i(0))^2 + (z_i(t) - z_i(0))^2]
\]

(1)

The key transport property called diffusion tortuosity \( \tau \) of the porous medium is related to the time-derivative of \( <r(t)^2> \) and can be expressed as:

\[
\tau = \frac{A}{d < r(t)^2 > / dt} \quad \text{as} \quad t \to \infty \quad (2)
\]

where \( A \) is a constant depended of implemented image lattice parameters and by some authors \([7,8]\) is assumed as 1.

The way of determining of \( \tau \) as a converging limit of \( d <r(\tau)^2>/dt \) needs the explanation. As time \( t \) elapsed random walkers migrated in porous media further than the average pore size. In the long - time limit they experienced the tortuosity of the surrounding material and the slope of \( <r(\tau)^2> \) reached a constant value. Hence, the tortuosity is defined by (2). In practical condition to determine the slope \( d<r(\tau)^2>/dt \) a one uses a procedure performing the calculation of the statistical coefficient of a least-squares linear regression using the consecutive 100 values of \( <r(\tau)^2> \) taken for the large values of \( t \).

According to the assumptions found in \([8]\) the authors of present paper have tested 6000 walkers and the slope of \( d<r(\tau)^2>/dt \) was checked in the proximity of \( t = 400,000 \) (i.e. for \( t = 399100, 399101 \) and so on...) for ten times. The resulting value of diffusive tortuosity was calculated as the average of the obtained results for which the correlation coefficient of the linear regression procedure was greater than 0.8. The diffusive tortuosity is an idirec function of the porosity of the body. The Fig. 2. below presents two model structures characterised by the volume fraction of porosity equalled 1%. The porosity is represented by the rectangular channel crossing the central part of both structures. At the left side of the figure the channel possesses continuing interior and at the right side the channel is divided approx. into two halves by the thin obstacle. The tortuosity calculated with the program described in present paragraph resulted in a value of 28 in the case of the long channel and in a value of 56 for the obstacled variant, respectively.

Three concrete mixes were designed and prepared of the same ingredients but with different water to cement ratio to result in different level of porosity. Sand of fraction 0-2 mm as fine aggregate and crushed amphibolite of fractions 2-8 and 8-16 mm as coarse aggregate were used. The composition of concrete mixes is shown in Table 1.

The beams 100×100×500 mm were cast for testing of progress of carbonation and stored for 27 days in water at temperature of +22°C. Then they were stored at laboratory conditions at +20°C, 60% RH until the mass equilibrium was reached (14 days).

Micro-CT method was applied with Nanotom 30 microtomograph made by General Electric operating in the Institute of Materials and Machine Mechanics in Bratislava. The space resolution of the reconstructed microstructure was 5 \( \mu m \) per voxel.

| Mix designation | cement CEM I 42.5 R | fine aggregate 0-2 mm | coarse aggregate 2-8 mm | coarse aggregate 8-16 mm | water | superplasticizer, % cement mass | Mix specific density [kg/m³] |
|-----------------|---------------------|-----------------------|-------------------------|-------------------------|------|-----------------------------|-----------------------------|
| A               | 350                 | 630                   | 695                     | 655                     | 158  | 0.30                        | 2424                        |
| B               | 400                 | 580                   | 625                     | 615                     | 200  | 0.00                        | 2433                        |
| C               | 320                 | 630                   | 675                     | 650                     | 176  | 0.00                        | 2431                        |
The result of micro-CT scanning was a set of tomograms (specimen cross-sections), performed every 5 µm along the specimen height. The digitized form of tomograms characterized the microstructure of specimens and included the information about the material in form of values of brightness in arbitrary units [a.u.] for all pixels of extracted specimen piece. Voxel brightness was proportional to the local density of the material. Two data subsets (ROIs) were extracted from the datasets representing the specimens. The ROIs labelled A1, B1 and C1 characterized with cement matrix to aggregate ratio of ca. 40% are presented of Fig. 3.

To determine the impact of cement matrix amount to the results of calculating of tortuosity, the other group, labelled A2, B2 and C2 was prepared in such a way to obtain cement matrix to aggregate ratio of ca. 30% what is depicted in Fig. 4.

In order to determine magnitudes of brightness of voxels belonging to pores, cement matrix and aggregates greyscale value histograms were performed for ROIs presented in Figs 3. and 4. These histograms are presented in Fig. 5.

In a part of the histograms presented in Fig. 5 one can mention distinct discontinuities at histogram curves resulted by the transition from one material phase to another (i.e. pores->matrix-> aggregates) what was discussed in [8]. It is obvious that the magnitude of the discontinuities depends on the volume distribution of phases in investigated piece of the material. Assuming the information which was derived from the greyscale histograms presented above the authors of the paper decided to take the following average brightness levels to distinguish the phases within analysed ROIs: 40 as the pores-matrix transition and 70 as the matrix-aggregates transition. It was proven that the variations made on these levels in magnitude of 2 or 4 percent have little influence for further determination of microstructural parameters.

After the determination of greyscale levels related to three principal material phases it was possible to process the prepared ROIs using the algorithm of random walk presented in previous chapter. The graphical illustration of the range of walkers migration in the ROIs od type 1. is presented in Fig. 6. The visualisation was made after 400,000 of time steps. The final positions of first 500 walkers are marked with small dots.

| material parameter                                      | specimen A | specimen B | specimen C |
|---------------------------------------------------------|------------|------------|------------|
| water to cement ratio                                   | 0.45       | 0.5        | 0.55       |
| pore volume to matrix volume in specimen part characterized with cement matrix to aggregate ratio of ca. 40% | 0.8 %      | 4 %        | 18 %       |
| pore volume to matrix volume in specimen part characterized with cement matrix to aggregate ratio of ca. 30% | 0.8 %      | 2 %        | 17 %       |
| diffusive tortuosity in specimen part characterized with cement matrix to aggregate ratio of ca. 40%  | 282        | 43         | 81         |
| diffusive tortuosity in specimen part characterized with cement matrix to aggregate ratio of ca. 40%  | 244        | 96         | 46         |

For each ROI the ratio of pore volume to matrix volume was determined. The latter parameter called 'segmented
porosity’ can be used to characterize the concrete microstructure. The results of the analysis of the data obtained from the specimens by means of micro-CT method are summarized in Table 2.

4. Conclusions

The results of the investigation allow the authors to conclude that the micro-CT method is capable to deliver some information on the microstructure of tested concrete specimens after contactless and relatively fast inspection. The important point of the data analysis is the proper determination of the pores-matrix threshold and matrix-aggregates threshold in respect to the population of examined voxel brightness parameter. After the assumption of the latter parameter it is possible to determine the segmented porosity what is proportional to water to cement ratio and thus is proportional to the material porosity. It is also possible to calculate the diffusive tortuosity - the parameter describing the pore connectivity of the investigated material. However that parameter presents non-linear dependence on segmented porosity as it was discussed in [8] because it is sensitive to the configuration of pore system. The authors of this paper have determined the close related results of diffusive porosity for specimens with w/c parameter of 0.5 and 0.45 but this parameter was of significant different magnitude for specimen with higher w/c ratio. The procedure of determining of diffusive porosity resulted in a relatively large deviation of results and perhaps need more repetitions. However it is evident that combined with the derived value of segmented porosity can be a fast tool for determining of microstructural parameters of concrete in some applications.

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