Dependencies between Cracking Patterns and the Physico-Mechanical Properties of Microsilica Modified Cement Matrix

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Abstract. The article presents and evaluates the dependencies that occur between the parameters describing the geometry of cracking patterns on the sample's surface and the selected physico-mechanical properties of cement pastes with the addition of microsilica. The stereological parameters obtained with the use of image analysis were applied to the quantitative description of cracking patterns; also the compressive and tensile strength as well as the apparent density were examined. Cracks analyzed in the surface were caused by loading the samples with an elevated temperature. The relationships were calculated using the least squares method, and the quality of the fit of the model curves to the experimental data was evaluated on the basis of the diagnostic statistics. The results obtained indicated the existence of very strong relations between the geometry of cracking patterns and selected material features of the cement matrix. The analysis carried out is also of a great practical importance as it can help in assessing the degree of degradation of a cement structure exposed to elevated temperatures.

1. Introduction
As a result of the temperature gradient, the cement matrix may crack. Cracks through propagation combine or intersect to form a characteristic cracking pattern, which in the literature is also referred as the cluster cracks/thermal cracks/mapcracking [1, 2]. There are only few research papers that attempt to quantitatively analyze cracking patterns because it is difficult from a methodological point of view. However, such analyzes have a high scientific and practical value because cracks of the cement matrix increase the risk of penetration of harmful substances into the material, causing its progressive degradation. Currently, the most commonly used concept to describe cracking patterns of cementitious materials is the crack density [3-5]. However, this is a very general parameter that does not provide detailed information on, e.g., the average crack width or the average area limited by cracks.

The authors of this work, to evaluate cracking patterns use image analysis tools, the advantage of which is that they are non-destructive methods. Their potential in the technology of cementitious materials is still not fully understood. The purpose of the research conducted was to determine whether, based on the knowledge of the geometrical parameters of the cracking patterns, it is possible to estimate the physical and mechanical properties of cement pastes with the addition of microsilica (MS). So far, no similar studies have been reported in the literature. For the purpose, a statistical analysis tool – the least squares method (LSM) was used. The quality of the dependencies calculated has been verified using the diagnostic statistics, i.e., the correlation coefficient, the determination coefficient, the standard error of estimation, and the coefficient of random variation. It was determined which...
physical and mechanical parameters of the cement matrix can be estimated with a high accuracy based on the measurement of geometric features of the cracking patterns.

2. Materials and Methods

2.1. Preparation of Modified Cement Matrix and Thermal Loading

Two series of cement pastes were tested, in which microsilica was used as a 10% substitute for cement mass. For both series, samples were made with three w/b (water/binder) indicators equal to 0.4, 0.5, and 0.6, respectively. The binder is understood as the total content of cement and microsilica in the material. The series differed from each other with the class of cement used; in the first series (C42MS), the ordinary Portland cement (OPC) CEM I 42.5R was used, and in the second series (C52MS), the OPC CEM I 52.5R was used. The cements used have a very similar chemical composition, which was presented in [6]. They differ in terms of grains' size; CEM I 52.5 R has a specific surface area greater by approximately 15% compared to CEM I 42.5 R.

Samples were made as bars with dimensions 40x40x160 mm³. They were made in accordance with EN 196-1 [7]; maturation took place in air conditions for 28 days (average relative humidity – 50%, average temperature – 22 °C). The samples were then subjected to an elevated temperature reaction based on the thermal shock. The samples were placed in a preheated oven at 250 °C for a period of 4 hours. The method of thermal loading was to extract the defects of the structure in the form of cracks, without excessive deterioration of the mechanical properties of the cement matrix. At this temperature, the cement matrix is thermally and chemically stable. The formation of cracks in the material is caused by the pressure of the saturated water vapor, which is associated with the intensive evaporation of free water. Thermal load parameters were selected based on previous studies [1, 8], in which cracking patterns were analyzed in the context of the self-assembly process of the cement matrix structure.

2.2. Properties of Modified Cement Paste

The bending tensile strength (f_{ct}) according to the EN 12390-5 [9], compressive strength (f_{c}) according to the EN 12390-3 [10], and the apparent density (D) according to the EN 12390-7 [11] was tested. The results of f_{ct} and D are the arithmetic means of 6 samples, in the case of f_{c} from 12 samples. The difference in number of samples was due to the fact that the f_{c} test was carried out on the halves of the beams obtained due to the f_{ct} measurement.

To quantify the cracking patterns geometry, stereological parameters obtained from the image analysis were used, which were measured using ImageJ v. 1.50d software. The scanned surface of the samples was analyzed. A measurement procedure was applied, which was described in detail in [8]. Among the stereological parameters, the average cluster area (A), the average cluster perimeter (L), and the average crack width (I) were measured. A cluster is understood as the area on the surface of the sample, which is limited on each side with a crack or edge of the sample. An example of a cracking pattern on the surface of a cement paste with the addition of microsilica is shown in Figure 1.

The results of the tests are presented in Table 1. Index (R) means the value obtained on the standard samples; index (T) – parameters for samples after temperature load.

![Figure 1. Cracking pattern on the surface of cement paste of C42MS series with w/b = 0.6](image-url)
Table 2 shows the calculated equations of curves together with the values of the diagnostic statistics random variation (i.e., the determination coefficient ($R^2$)). The adjustment of the curves calculated to the empirical data was checked using the diagnostic statistics, mechanical features was limited to one of the above mentioned parameters, i.e., each other. Thus, knowing the value of one parameter with very high accuracy, the value of the second which is a rare situation. However, this difference is very small and equal only to 0.02. In the series have obtained values more favorable than global coefficients. Values of dependencies were graphically shown in Figure 2 – Figure 4. The relationships for each series were calculated separately because in this situation a better degree of matching the model curves to the distances of all empirical points from the corresponding points on the fit line was as small as possible [12]. The dependencies relate to the condition of the material after temperature load. In the previous studies, it was noted that there is a very strong correlation (0.98-0.99) between dependencies; [1, 8, 13]. The equation was calculated, which makes these parameters to be dependent from variables. Assuming the level of the statistical significance $\alpha$ at 0.05, all $p$-values assume a smaller value. This means that the correlations occurring between variables, both for individual series and for the whole dependency, are in each case statistically significant.

2.3. The Least Squares Method

The dependencies that occur between the geometry of cracking patterns and the physico-mechanical parameters of the modified cement pastes were calculated using the least squares method (LSM). This method consists in such a selection of the curve equation, so that the size being the sum of the squares parameters of the modified cement pastes were calculated using the least squares method (LSM). This method consists in such a selection of the curve equation, so that the size being the sum of the squares parameters of the modified cement pastes were calculated using the least squares method (LSM). This method consists in such a selection of the curve equation, so that the size being the sum of the squares parameters of the modified cement pastes were calculated using the least squares method (LSM). The equation was calculated

$$\sum (y_i - \hat{y}_i)^2 \rightarrow \min$$

where $y_i$ is the $i$th observed value and $\hat{y}_i$ is the $i$th predicted value. The equation of the line is determined by minimizing the sum of the squares of the residuals, which is the difference between the observed and predicted values.

$$\hat{y} = \beta_0 + \beta_1 x$$

where $\beta_0$ is the intercept and $\beta_1$ is the slope of the line. The values of $\beta_0$ and $\beta_1$ are calculated using the following formulas:

$$\beta_1 = \frac{\sum (x_i - \bar{x})(y_i - \bar{y})}{\sum (x_i - \bar{x})^2}$$

$$\beta_0 = \bar{y} - \beta_1 \bar{x}$$

where $\bar{x}$ and $\bar{y}$ are the means of the $x$ and $y$ values, respectively.

3. Results and Discussion

Table 2 shows the calculated equations of curves together with the values of the diagnostic statistics that describe the following relationships: $f_{c(T)}(A)$, $f_{c(T)}(\bar{I})$, $f_{c(T)}(\bar{A})$, $f_{c(T)}(\bar{I})$, $D_{c(T)}(A)$, and $D_{c(T)}(I)$. Dependencies were graphically shown in Figure 2 – Figure 4. The relationships for each series were calculated separately because in this situation a better degree of matching the model curves to the experimental data was obtained; the correlation coefficients, in the criterion of division into individual series have obtained values more favorable than global coefficients. Values of $\rho$ are negative; this means that the values of the physico-mechanical parameters of the MS modified cement paste decrease with the increase of $\bar{A}$ and $\bar{I}$. Thus, the fewer clusters on the surface of the cement matrix, the less its strength. However, the key aspect in this case is that at the same time the crack width increases, which is the main reason for the weakening of the material structure.

Assuming the level of the statistical significance $\alpha$ at 0.05, all $p$-values assume a smaller value. This means that the correlations occurring between variables, both for individual series and for the whole dependency, are in each case statistically significant.

In global terms, the strongest correlation is characterized by the $f_{c(T)}(\bar{A})$ and $D_{c(T)}(\bar{I})$ dependencies; $\rho$ reach a very high value of -0.93 and -0.90, respectively. The interesting thing is that in the division into the series for $f_{c(T)}(\bar{A})$ the correlation coefficients assume lower values than in the global approach, which is a rare situation. However, this difference is very small and equal only to 0.02. In the remaining situations $\rho$ divided into series, achieves values greater than global coefficients. The weakest global correlation occurs for dependencies with $f_{c(T)}$. This is due to the fact that the C52MS
series was characterized by lower tensile strength than C42MS, while at the same time lower values of $A$ and $I$. However, in the criterion for division into series, the $\rho$ values are very high and show a very strong correlation.

The curves calculated best reflect the empirical data in a situation when the $A$ is variable, because $R^2$ assumes higher values (0.84-0.91). The quality of curve fitting for $I$ is still high (0.78-0.88), however, lower compared to the first variable. It is worth noting that for cement paste made of CEM I 52.5R, the $W$ values in each case are smaller than for the C42MS series. This means that the estimation of material values for the paste made of cement with a higher fineness will be characterized by greater accuracy than for samples made of CEM I 42.5R.

The coefficient of random variation reaches the lowest values for the relationship with $D(T)$. These values are even 6-7 times lower than the highest ones. Considering the high value of the determination coefficient, it can be concluded that the physico-mechanical properties of the cement matrix will be able to estimate most accurately on the basis of apparent density, both on the basis of $\bar{A}$ and $\bar{I}$.

### Table 2. Equations describing the relationships between the cracking patterns geometry and the physico-mechanical properties of cement pastes modified with microsilica.

| Relationship | Series | Equation | $R^2$ [-] | $S_e$ [MPa] | $W$ [%] | $\rho$ [-] | $p$-value (the $\rho$ significance test) [-] |
|--------------|--------|----------|-----------|-------------|---------|----------|----------------------------------|
| $f_c(T)(\bar{A})$ | C42MS | $y=5074x^{-1.130}$ | 0.84 | 4.78 | 17.50 | -0.93 | -0.91 | <0.001 | <0.001 |
| | C52MS | $y=1086x^{-0.775}$ | 0.91 | 3.89 | 9.86 | -0.91 | -0.91 | <0.001 | <0.001 |
| | C42MS | $y=0.063x^{-1.786}$ | 0.79 | 5.50 | 20.15 | -0.83 | -0.88 | <0.001 | <0.001 |
| | C52MS | $y=0.582x^{-1.228}$ | 0.78 | 5.90 | 14.94 | -0.83 | -0.82 | <0.001 | <0.001 |
| $f_c(T)(\bar{I})$ | C42MS | $y=199,18x^{-0.955}$ | 0.87 | 0.32 | 13.47 | -0.47 | -0.95 | 0.036 | <0.001 |
| | C52MS | $y=12,81x^{-0.474}$ | 0.88 | 0.12 | 7.08 | -0.47 | -0.92 | <0.001 | <0.001 |
| $f_{cf}(T)(\bar{A})$ | C42MS | $y=0.012x^{-1.565}$ | 0.88 | 0.30 | 12.72 | -0.71 | -0.96 | <0.001 | <0.001 |
| | C52MS | $y=0.103x^{-0.815}$ | 0.82 | 0.14 | 8.58 | -0.71 | -0.93 | <0.001 | <0.001 |
| $D(T)(\bar{A})$ | C42MS | $y=4,427x^{0.256}$ | 0.85 | 0.051 | 3.78 | -0.87 | -0.92 | <0.001 | <0.001 |
| | C52MS | $y=3,600x^{0.225}$ | 0.90 | 0.042 | 3.06 | -0.87 | -0.90 | <0.001 | <0.001 |
| $D(T)(\bar{I})$ | C42MS | $y=0.347x^{0.400}$ | 0.80 | 0.058 | 4.35 | -0.90 | -0.89 | <0.001 | <0.001 |
| | C52MS | $y=0.360x^{0.390}$ | 0.81 | 0.057 | 4.16 | -0.90 | -0.91 | <0.001 | <0.001 |

**Figure 2.** Relationship between $f_{cf}(T)$ and: a) $\bar{A}$; b) $\bar{I}$
Figure 3. Relationship between $f_{c(f)}$ and: a) $A$; b) $I$

Figure 4. Relationship between $D_{(f)}$ and: a) $A$; b) $I$

4. Conclusions

The article specifies the relationships between the cracking patterns geometry and selected physical and mechanical properties of cement pastes modified with microsilica. On the basis of the analyzed and interpreted research results, final conclusions were formulated:

- With the increase in the $A$ and $I$ values, all the examined physical and mechanical features of the cement matrix are reduced.
- The method of the least squares supplemented with the evaluation of diagnostic statistics is an effective tool in defining dependencies that occur between variables.
- The dependencies between physico-mechanical features and the cracking patterns geometry, in the criterion of division into series, are characterized by a very strong correlation (less than
In the case of global correlation, only in the case of the relation with $f_{cf}(T)$, the average correlation was achieved (-0.47 and -0.71).

- $D(T)f$ and $D(T)f^2$ are the relationships for which the highest fit of the model curves has been achieved ($R^2$ from 0.80 to 0.90). In addition, the lowest value of the random coefficient of variation (3.06-4.35) indicates that the apparent density of the cement matrix modified with microsilica can be estimated with very high accuracy based on the geometric characteristics of the cracking patterns.

- The assessment of physical and mechanical parameters of cement pastes based on the measurement of parameters obtained from the image analysis is very valuable, because it is a non-destructive test. This is of great practical importance, because the tests conducted have proved that by measuring parameters of the cracking patterns such as $\bar{A}$, $\bar{L}$ and $\bar{I}$, it is possible to estimate the value of selected material properties of the cement matrix with high accuracy. It may improve the process of analysis of the degree of damage to the cement structure due to the influence of an elevated temperature.

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