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Methodology of discovering and control of clay roofing tile textural inhomogenities

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Abstract

The adjustment of shaping parameters, including vacuum press working ones, is an important factor in the production of ceramic roofing tiles, especially regarding their textural characteristics. This paper is focused on the investigation of the screw conveyer wearing degree of the vacuum press in the period of 13 weeks, with the aim to define its time of replacement. The industrial clayey raw material with a considerable amount of quartz, orthoclase, mica and traces of carbonates presented the composition used in the tile manufacturing process. A control method which correlates the textural characteristics of tile segments with the degree of screw conveyer wearing was set up. This method could be a useful tool in the prevention of tile lamination and small pores appearance regarding the extruded column shaping by a vacuum press.

Keywords: Extrusion; Defects; Thermal expansion; Clays; Functional applications

1. Introduction

The final characteristics of ceramic roofing tiles, reflecting inter alia the textural inhomogenities, are the results of raw material characteristics, processing parameters and mechanic-technical parameters.[1] Regarding processing parameters, the textural inhomogenities could be the result of relatively complex flow processes which occur inside the extruder during the shaping process. In fact very slight changes in the plasticity of clay mass could influence the pore structure design and initiate roofing tile shaping defects (lamination, roughness of the column, "dragon's teeth"). On the other hand, it is evident that the presence of internal inhomogenities is caused by the existence of different exit speeds of the clay mass during the extrusion procedure. This effect forces the clay mass to change its orientation and causes the formation of the extruded column parts of different densities. The characteristics of the worn screw conveyer of vacuum press could produce the changes of mass plasticity. The determination of the moment of its replacement in order to maintain the final characteristics of ceramic roofing tiles at appropriate level would be of great practical interest.

In previous investigation[2] it was shown that the development of textural inhomogenities, in some parts of roofing tiles, was due to the existence of inadequate shaping procedure parameters. It was noticed that by alternating the operating shaping parameters, the vacuum values and speed of extruded clay column, the amount of the critical pore diameter of roofing tiles could be minimized. These values were linked with the sample linear dilatation values, within the temperature interval −40 °C to +40 °C, for defining the volume of ice which could be formed in the pores of specified characteristics. The moment when the values of operating shaping parameters have to be changed was not determined.

The present paper is dedicated to the time definition of the replacement of the worn screw conveyer of vacuum press in order to manage to avoid the formation of internal textural inhomogenities in the extruder column. Besides common methods for the characterization of internal textural inhomogenities, water absorption and diametrical compression values, relative dilatation changes of the tested samples were used as the relevant indicator of the textural inhomogenities. An attempt has been made to develop one cycle freezing/thawing testing method

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for the prediction of textural inhomogenities of ceramic roofing tiles. Many factors and their mutual relations, at low temperatures, play an important role in the roofing tile relative length changes: permeability parameters of the analyzed system, cooling rate during freezing, saturation degree and migration of water. The amount of nonfreezing water in such cooling conditions,[3] which is always greater in freezing than in thawing procedure, also plays important role. Regarding our experiments, the freezing test was conducted with fast cooling rate in order to create more severe testing conditions. The freezing procedure in dried conditions[4] provided information about the existence of inhomogenities in phase composition, while the water saturated conditions emphasized the importance of supercooling effects of water.[5] It is known that supercooling conditions give the possibility of the appearance of damages due to the hydraulic pressure increase, as well as the water diffusion existence through the water layers[3] of wall pores. The mentioned consequences of the fast cooling rate, in the case of our investigation, influenced the relative dilatation values of the samples which, correlated with the pores characteristics (size pore values, pore specific surface values and pore specific volume values), have given an accelerated methodology of discovering and control of clay roofing tile textural inhomogenities.

2. Experimental

The standard industrial manufacturing line of ceramic roofing tile production was used for the production of 13 series of ceramic tiles whose extruded column was formed with the screw conveyer of defined characteristics (life span), Fig. 1. Each series consisted of eleven roofing tiles sampled during the period of one working week time. The total time of investigation was 13 weeks. Each tile was divided into 24 segments (in total 3432 segments). All segments of the first six roofing tiles of each series (in total 1872 segments) were the subject of water absorption characterization, EN 532/2-1998, water saturation value, ASTM C-373 (936 segments) and diametral compression values, Instron 1122 Press—crosshead speed of 0.2 mm/min (the other 936 segments). Based on the used statistical methods,[1] Fuzzy C-means, Fuzzy learning vector quantization, Fig. 2, three series of different screw conveyer life times were selected: series III (beginning), VIII (middle) and XII (end) of the screw conveyer life span. The other 5 tiles, from the total number of 11 tiles of the above selected three series of tiles, were used for the assortment of the segments of the expressed textural inhomogenities, notified as 9, 12, and 21, Fig. 1. These segments were the subject of textural characterization, freezing dilatation measurements and microstructure investigation.

Fig. 1. Schematic presentation of the roofing tile design with the selected tile segments.

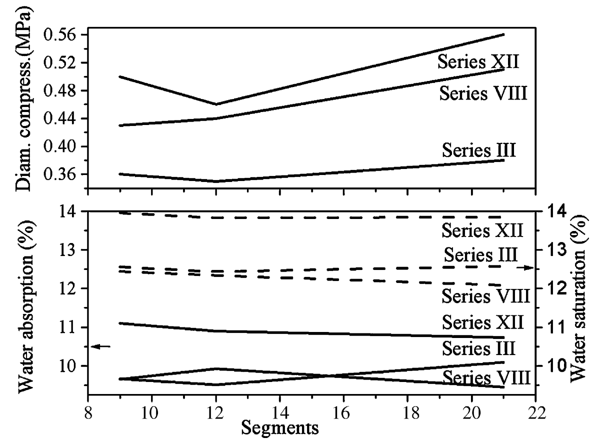

Fig. 2. Water absorption, water saturation and diametral compression values of the 13 series of ceramic roofing tiles.

The raw material used for the tiles production was the clayey material with a considerable amount of quartz, orthoclase, mica and traces of carbonates.

For the textural features characterization of the samples segment, the mercury intrusion porosimeter (Model 2000, Porosimeter WS, Carlo Erba, Rodano, Milano, Italy) was applied, while a low temperature nitrogen adsorption (Model 2000, ASAP Micrometrics, USA) was used as the method for the evolution of specific surface area and for the characterization of pores whose diameter was less than 0.1 μm. The SEM observations (Model JSM–6460HV, JEOL, Tokyo, Japan) were performed on the specimens series treated in water saturated conditions (24 h), later cured 24 h in the refrigerator (−18 °C). The microstructure of the fresh fracture, coated with gold, was analyzed "in situ". Sliced specimens (size 4 mm × 4 mm × 2 mm) were cut from the selected segments for the low temperature dilatation measurements (Model 981, Thermo Mechanical Analyzer, Du Pont, Delaware, USA), then ground with a 400 grit SiC paper for obtaining parallel surfaces. Before the application of the dilatation measurements the specimens were quenched in liquid nitrogen. After the abrupt decrease of the temperature down to −40 °C the analysis of the sample thawing behavior was started. The temperature interval of the investigation was from −40 °C up to +40 °C, with a rate of thawing of 10 °C/min.

Table 1
Total porosity, specific surface area of pores of the analyzed segments and the maximum difference between the relative dilatation (water saturated and dry conditions), points C and D

Series	Segments	Total porosity (%)	Specific surface area of pores (m^2/g)	$\Delta(\Delta L/L_0)_{max} \times 10^{-4}$ between points C and D
III	9	32.39	0.7816	2.54
	12	33.41	0.7437	3.86
	21	32.02	0.872	3.37
VIII	9	32.57	1.1981	6.01
	12	33.01	0.7832	1.13
	21	32.71	0.8629	1.76
XII	9	33.12	0.139	2.72
	12	36.96	0.9891	4.3
	21	33.11	1.0529	5.74

These measurements were performed with dried and 24 h water pretreated (submerged) samples (T_{water} = 21 °C).

3. Results and discussion

Water absorption, water saturation values and diametral compression values: The values of water absorption, water saturation and diametral compression, obtained as the average value of the specified segments of three roofing tiles, are presented in the Fig. 2. The series XII of the tiles, presents the series of the highest value of water absorption, water saturation and diametral compression values, but also the series of the smallest differences between segments values and highest degree of homogenization (water absorption and water saturation values). This feature expresses the strength of cohesion forces inside the material layers. Evidently, the results of the three types of investigation only point to the existence of textural inhomogenities.

3.1. Textural characteristics investigation

Hg porosimetry measurements: Among the total porosity values of the selected tile segments of the series III and VIII, Table 1, no distinguished differences could be noticed. This fact is not valid in the case of the segments of the series XII. The segment 12, of this series, have value of the total porosity which is ~12% larger than the values of the other two segments. In view of the pore size distribution of the segments, the series III possesses the most homogenized distribution, Fig. 3. This series could be characterized as the system whose dominant pore radius range from 0.5 to 2.0 μm. The series VIII possesses the same broad dominant pore interval, 0.5 to 2.0 μm, while in the case of tile series XII, it is shifted towards the narrower pore radius interval, 0.5 to 1.0 μm. An evident increase in the macrocapillary porosity for this series is also identified,[6] the fact which is in good correlation with the water absorption values of the tile segments of this series.

Low temperature nitrogen adsorption measurements (*pores of diameter* <0.1 *μm*): The tile segments of the series III and VIII could be characterized as the ceramic systems with a developed mesopore structure, whereas the tile segments of the series XII possesses additional micro pores (pore diameter ~0.008 μm), Fig. 4. The segment 12 of the series III and VIII could be characterized as the one which performed a pore diameter shifting of the pore volume maximum. This fact characterized the segment 12 as the most unstable among the three analyzed segments. The

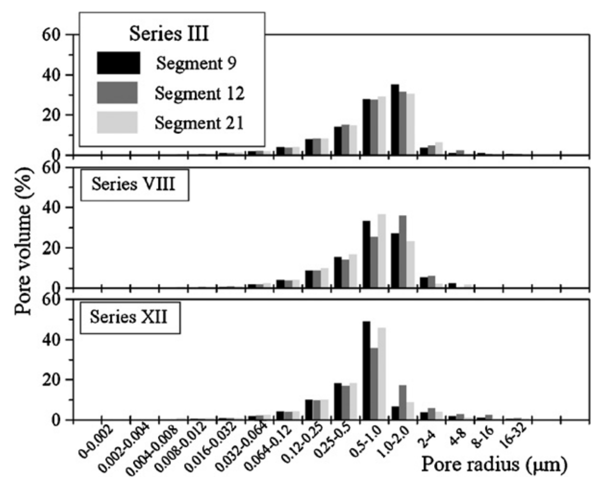

Fig. 3. Pore size distribution (Hg porosimetry) of the selected tile segments of the series III, VIII and XII.

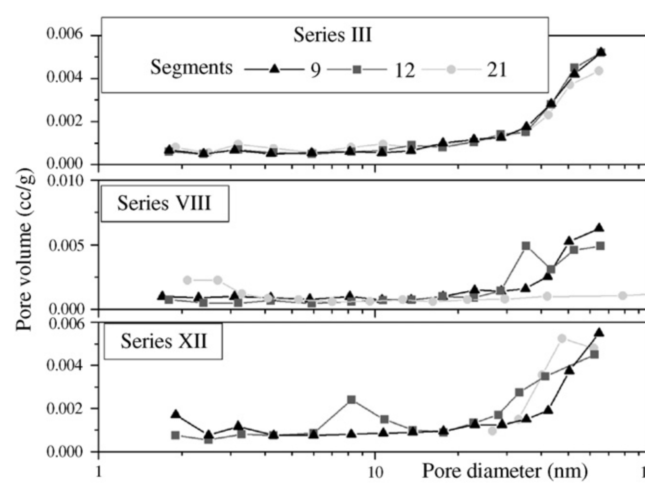

Fig. 4. Pore size distribution of the selected tile segments of III, VIII and XII tile series (low temperature nitrogen adsorption).

Table 2
Specific surface area, pore volume and pore diameter values

Segments	Series III			Series VIII			Series XII		
	9	12	21	9	12	21	9	12	21
Specific surface area (m^2/g)	1.367	1.221	1.138	1.8312	1.2151	1.22	0.7037	1.049	1.5
Specific surface area of pores (m^2/g)	0.7816	0.7437	0.872	1.1981	0.7832	0.8629	0.139	0.9891	1.0529
Pore volume, exp −2 (m^3/g)	0.2274	0.2271	0.2292	0.2967	0.2329	0.2243	0.1661	0.2973	0.2583
Pore diameter (nm)	4.9553	5.5391	6.617	4.91	5.48	6.173	4.678	8.35	5.82

same segment has already been marked, Table 1, as the particular one regarding the total porosity value.

Among the segments of the series III a uniform pore size distribution could be noticed. This aspect of the pore size homogenization is also confirmed by analyzing the values of specific surface area of the materials, specific surface area of pores, specific pore volume values and pore diameter values, Table 2.

3.2. Low temperature dilatation investigation

The interpretation of the low temperature dilatation curves recorded in this test, Fig. 5, are the result of superimposure of several physical phenomena. For example, the curve obtained in dried conditions of investigation reflects the relation between the total porosity value and phase composition of the tile segments. A lower relative dilatation value present, generally for heavy clay products, the consequence of a greater amount of crystal amorphous phase or greater total porosity value or the sum of the both facts.[4] For the water saturated samples all the above-mentioned phenomena are additionally increased, due to the existence of the ice pressure on the pores wall, or the presence of an significant quantity of unfrozen water.

Dried conditions of investigation: Concerning small differences among the values of the total porosity, Table 1 (except the case of the segment 12-series XII), the phase composition of the tile segments could be taken as the influencing factor of the relative dilatation values obtained in the dried conditions of the investigation. The differences among the relative linear dilatation values of these segments could be explained as the consequence of the phase composition inhomogenity of the three series, Fig. 5.

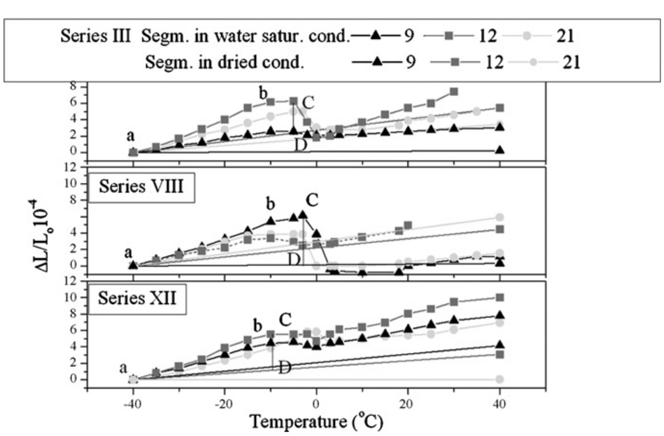

Fig. 5. Low temperature relative dilatation values for the series III, VIII, XII-dried and water saturated freezing conditions.

Water saturated conditions of investigation: The first part of the curves, Fig. 5, during the thawing procedure of the water saturated samples is practically linear (regions a and b). The value of each point of this region corresponds to the expansiveness value of the sample at the define temperature. In the point “b” the amount of the ice that changed its phase state, induce a decrease in the linear expansion of the sample. The developed inner stresses, up to the point C, could be related to the relative dilatation values of the sample in the dried freezing conditions (C and D), Table 1. Calculated as the difference between the two points (C and D), Fig. 5, this value represents the maximum of the sample dilatation value obtained in the thawing procedure only due to the ice/unfrozen water presence. Matching this value with the specific surface area of the pores (one of tile segment textural characteristics) it is possible to recognize a good correlation between these two parameters considering the tile segments position as well as the ages of the selected series (III, VIII, XII). The highest degree of homogenization exists among the values of the tile segments of the series III, while the lowest in the case of the tile segments of the series XII. The existing correlation could be taken as the control method for the tile segments inhomogenities detecting the screw conveyer state during its working period. It seems that surface phenomenon of pore wall plays the crucial role in the determination of the relative linear dilatation in the case of fast cooling. More precious data are obtained based on the “in situ” microstructure investigation of the water saturated and frozen tile segments.

3.3. Microstructure investigation of tile segments

The subject of the microstructure investigation were only the tile segments of the series VIII. The choice was made based on the results of the textural characteristics investigation, Table 2, and the results of low temperature measurements, Fig. 5 and Table 1 (series VIII). Two groups of samples could be distinguished inside the selected roofing tile series. The segment 9 represents the first group, while the other two segments (12 and 21) the second one. The SEM observations were performed “in situ” on the water submerged specimens (24 h), later cured 24 h in the refrigerator (−18 °C). The state of the tile segments during thawing procedure was analyzed. It is observed that the macro capillary pores of the segment 9 are completed with the ice, detail A of the Fig. 6. The ice melting/sublimation process is starting in the pore center (detail in Figs. 6B and 7) pointing out

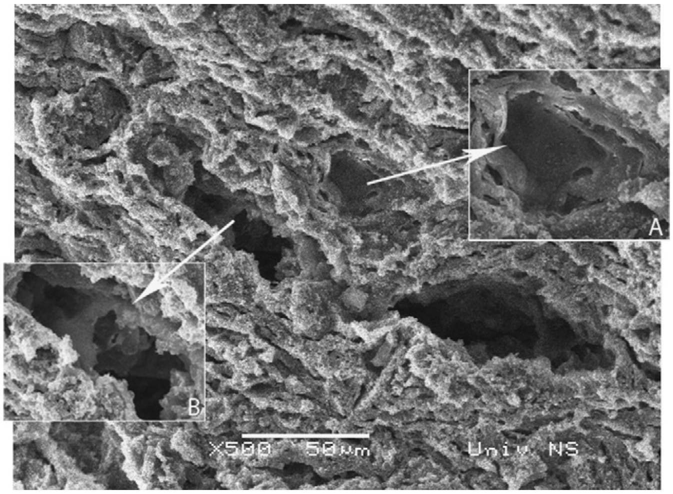

Fig. 6. SEM image of the segment 9, series VIII.

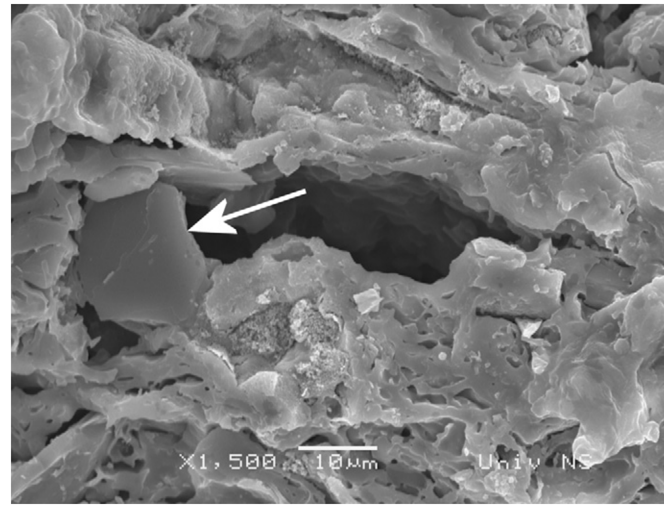

Fig. 9. SEM image of the segment 21, series VIII.

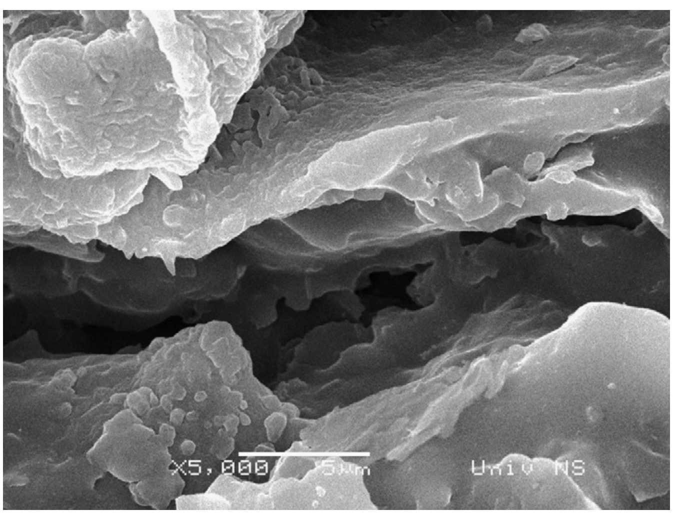

Fig. 7. SEM image of the segment 9, series VIII.

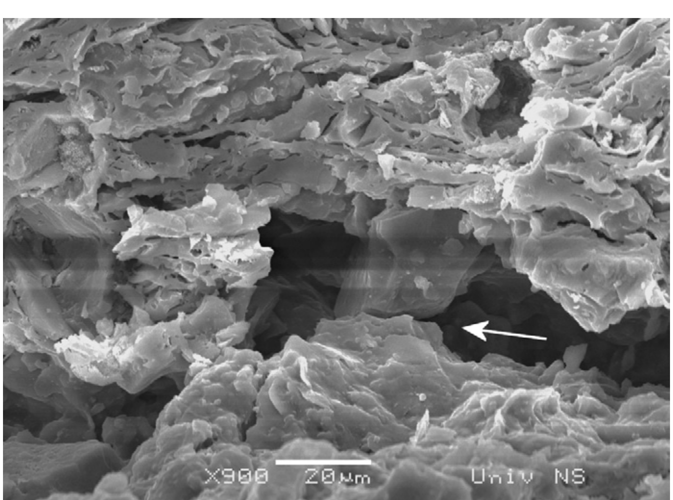

Fig. 8. SEM image of the segment 12, series VIII.

the extremely strong adhesion of the ice to the pore wall. The explanation could be the existence of high value of specific area of pores (high roughness of the pore surface), Table 2, which increase formation of nucleus at grain boundaries, as the result of the high degree of the ceramic body crystallinity. The segments 12 and 21 of the same series, could be characterized as the ones whose macro pores are only partially filled with the ice body, Figs. 8 and 9. The ice crack (see the arrow on the figures) is the consequence of the higher value of the ice adhesion to the pore wall than the tensile strength value of the lump of ice. Becoming free, in the water presence, the lump of ice obtains a great degree of mobility, and could cause the frost deterioration of the sample, Fig. 9 (segment 21/series VIII). Evidently the superimpose of the two phenomena, the ice action on the pore wall surface and the dilatation values (small C and D value), as a result of phase composition of the solid phases (crystal and amorphous phase) could have a serious consequence on the tile segment stability.

4. Conclusion

Data of water absorption, water saturation and diametral compression have detected the tile series of problematic textural characteristics. The additional investigated textural characteristics, total porosity values, specific surface area and specific pore volume values, diagnosed the location of the problematic segments. A good correlation between the surface area value of pores and the difference of the two maximum dilatation values (water saturated freezing conditions, dried freezing conditions) at the temperature of the ice melting (in the pores of the defined diameter) was set up. This correlation turns out to be a control method of tile textural inhomogenities regarding the worn-out level of the screw conveyer of the vacuum press. The “in situ” investigation of the microstructure of the water submerged specimens (24 h) later cured 24 h in the refrigerator (−18 °C) gave more detailed information about the ice/pore wall contacts in the tile segments of the specific textural characteristics giving a physical image to the control parameters of the established method.

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