ISSN 1392–1320 MATERIALS SCIENCE (MEDŽIAGOTYRA). Vol. 19, No. 4. 2013

The Effect of Firing Temperature on the Irreversible Expansion, Water Absorption and Pore Structure of a Brick Body During Freeze-Thaw Cycles

Mikuláš ŠVEDA [1] *, Radomír SOKOLÁŘ [2]

[1] *Faculty of Civil Engineering, Slovak University of Technology,, Radlinského 11, SK-813 68 Bratislava, Slovak Republic*
[2] *Faculty of Civil Engineering, Brno University of Technology, Veveři 331/95, CZ-602 00 Brno, Czech Republic*

crossref http://dx.doi.org/10.5755/j01.ms.19.4.2741

Received 31 October 2012; accepted 21 January 2013

The paper deals with the monitoring of brick body in the process of volumetric freezing and thawing. The samples were fired at temperatures of 900, 1000 and 1060 °C. Attention is focused on monitoring of the irreversible expansion, water absorption and pore structure of a brick body. We found that in all cases the endpoints take place continuously, where the amount firing temperature plays a crucial role. The greatest influence of freeze/thaw cycles on the change of the pore structure was also observed at the lowest temperature. The change of the pore system during the freeze-thaw cycles occurs in such a way, that the pore volume of small pores further decreases and conversely, the pore volume of large pores increases. The knowledge gained can be used not only in the production of new but also in predicting the remaining durability of older clay roofing tiles.

Keywords: brick body, clay roofing tile, frost resistance**,** irreversible expansion, water absorption, pore structure.

1. INTRODUCTION

An important property of clay roofing tiles or facing bricks is the frost resistance, which is considered as the resistance of a fired brick body in a moist condition against an alternating action of water and frost. In this case, we encounter three factors working together.

The first factor is the change in the state of matter from water to ice, it can cause a well-known expansion. The ice formation in porous material at the same time depends from the capillary diameter and extent of negative temperatures. With the decreasing diameter of capillary a negative temperature decreases and this relationship can be expressed by an exponential function [1]. The whole process of water freezing takes place in the form of rubble-ice in the larger capillaries being pressed into the smaller capillaries, the pressure increases and gradually leads to the formation of microcracks [2]. The size of the pressure in the smallest capillaries can achieve a value up to 200 MPa [3]

The second factor: the melting of ice leads to an increase of the expansion due to higher thermal linear expansion coefficient of ice as has the brick body itself. (thermal linear expansion coefficient of ice is $\alpha \approx 50\cdot10^{-6}$ K^{-1} and of brick body only $\alpha \approx 5{,}5\cdot10^{-6}$ K^{-1} [4]). The pressure from ice and melting ice applied on the pore walls causes an irreversible expansion of the brick body after each freeze-thaw cycle [5 – 7]. Concurrently a cumulative effect occurs until a total collapse is achieved [5, 8, 9].

The third factor: the moisture expansion of a brick body, which can induce reversible or irreversible volume changes [10 – 13]. The fact that the irreversible changes are of a larger dimension can pose serious problems especially for large scale products. The irreversible moisture expansion occurs mainly in brick raw material with a relatively high content of clay minerals fired at temperatures in the range about 600 °C to 1050 °C. It is a spontaneous process of the water reaction with the non-crystalline phase of the brick body, where the irreversible volume changes can usually exceed the value of 0.8 mm/m [13].

An intensive monitoring of the relationship between pore structure and frost resistance was carried out in the last 30 years out, where most attention was devoted to the distribution of the pores before the first freezing cycle [14 – 19]. Smaller attention is devoted to the distribution of pores after the end a certain number of freeze-thaw cycles, for example in the evaluation of old brick products (roofing tiles and bricks) [15, 20, 21].

It was found that the frost resistance of brick body can be better presented through the distribution of pores rather than just by its porosity, which is expressed through the pore volume or through the water absorption [22]. The actual distribution of pores is, however, influenced by several factors, such as firing temperature, mineralogical and granulometric composition of raw materials and the like [22 – 28].

Undoubtedly the most important impact has the firing temperature or the presence of a flux [29 – 33]. Insufficient firing temperature will have created a wide range of pores but it achieves only a low level of frost resistance [34]. Similarly, it is disadvantageous to have a very narrow spectrum of pores [35]. Many authors have tried to define most preferred composition of the pore structure, however neither proposal has been able to perfectly represent the

*Corresponding author. Tel.: +421-2-59274691; fax: +421-2-52494357.
E-mail address: *mikulas.sveda@stuba.sk* (M. Šveda)

relationship between the pore structure and frost resistance for different types of brick products [36 – 42].

As shown in the work of Raimondo et al. [41], a common feature of high frost resistance of a brick body on the basis of previous knowledge is a large number of pores larger than 3 mm. This requirement should satisfy the conclusions, as stated by Šveda [22, 29, 32]. The author has found with the various raw materials and at different firing temperatures that the higher value of the median pore radius, it is also higher frost resistance of brick body. The relationship between the median pore radius and frost resistance can be expressed by a mathematical function. This positive effect of the median pore radius on the frost resistance can see also in the works [30, 43].

So far, there has been a little studied area, which monitors the gradual changes in pore structure of a brick body namely during the freezing cycles. Therefore we can presume that the explanation of these changes could have a positive influence for example:

- in obtaining a high frost resistance brick product;
- in predicting the remaining durability of clay roofing tiles or facing bricks;
- in assessment the information about firing temperatures on the old roofing tiles or facing bricks.

2. EXPERIMENTAL

We have used the raw material from a plant, which produces roofing tiles. It is characterized by quaternary sediments, which are represented by layers of clay and silty loam soils of eolian origin. Its chemical and granulometric composition are shown in Table 1 and Figure 1. From the mineralogical point of view it is the montmorillonite/illite, almost without the calcium carbonates.

Table 1. Chemical composition of clay

Chemical composition, %	Clay
SiO_2	71.81
Al_2O_3	13.37
Fe_2O_3	5.28
CaO	0.46
MgO	0.91
Na_2O	0.45
K_2O	1.46
$CaCO_3$	0,63
$MgCO_3$	0.74
Loss on ignition	4.71

The test samples were prepared by cutting unfired roofing tiles lengthwise in three parts, which were brought from brickworks. They were consequently fired in an electric furnace at temperatures of 900, 1000 and 1060 °C. The firing scheme is shown in Figure 2. For the determination of an irreversible expansion we used the edge part of the beaver tail clay tile. For the determination of changes in the pore structure during freeze-thaw cycles we used only the middle part. For each firing temperature was used 5 pieces of beaver tail clay tiles.

The X-ray analysis of the samples (Fig. 3) fired at temperatures from 900 °C to 1060 °C in addition to the dominant quartz contains some mineral forms from the feldspar group, micaceous species and hematite.

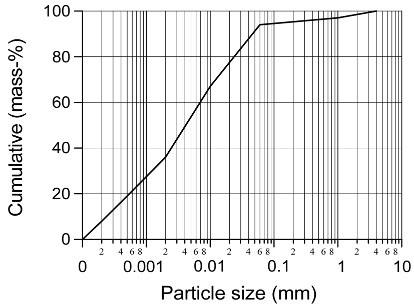

Fig. 1. Granulometric composition of clay

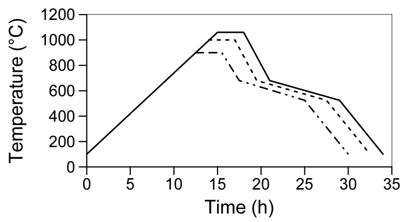

Fig. 2. Firing curve at temperatures of 900, 1000 and 1060 °C

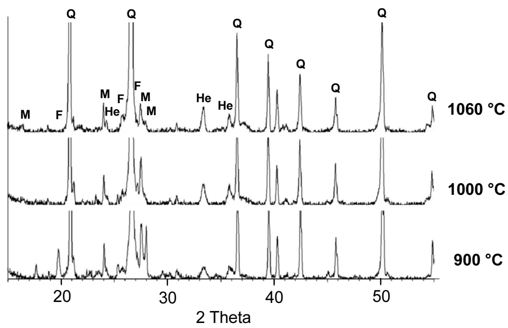

Fig. 3. X-ray diffraction profile for fired samples (Q = quartz; F = feldspar group; M = micaceous species; He = hematite)

3. TESTING PROCEDURES

Monitoring of changes in the brick body was realized in such a way so as to resemble the actual conditions in nature, i. e. brick products saturate water for a certain period of time and then they are subjected to freeze-thaw cycles. Our aim was therefore to monitor the change processes in the brick body. In the first phase the samples were placed in the water bath for 21 days and the irreversible moisture expansion and water absorption were monitored. In the second phase, i. e. during the freeze-thaw cycles, we have further studied the irreversible expansion, water absorption and the changes in pore structure of brick body (volume and median pore radius).

The irreversible expansion was determined with a deformeter, where the starting distance between two dots on a face of the test specimen was 200 mm. The measurement accuracy was 0.01 %.

Frost resistance of the saturated water samples was determined though standard freeze-thaw cycles: 20 hours at air temperature of –18 ±2 °C and 4 hours in water temperature of 18 ±2 °C, i. e. one freeze cycle was carried out in 24 hours.

The pore structure of the brick body was determined by high-pressure mercury porosimeter (Thermo Finnigan Pascal 240, firm Thermo Scientific). The test samples for this measurement during freeze/thaw cycles were gradually taken from the same batch.

4. RESULTS AND DISCUSSION

The course of irreversible moisture expansion at all three firing temperatures is shown in Figure 4. In the first ten days of measurement, a rapid increase in the expansion we observe at all three temperatures. The fastest increase was carried out at temperature of 900 °C, where the highest final values were also achieved

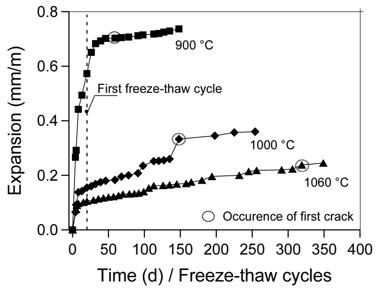

Fig. 4. Effect of deposition in water and freeze/thaw cycles on the irreversible expansion (typical course on one sample)

After a rapid increase in expansion follows a slowdown and it goes continually until the breach of the sample. In the firing temperatures of 1000 °C and 1060 °C is this slowing down observed even during storage in an aqueous medium, but at temperature of 900 °C up during the freeze/thaw cycles.

With increasing firing temperature there occurs a significant shift in creation of the first visible crack, see Fig. 4. Similar results as with irreversible expansion we obtained well as with water absorption. The values of irreversible expansion are in accordance with authors [13].

The highest absorption after 21 days storage in water was achieved at the firing temperature of 900 °C and its steady state occurs in a rather short time, see Figure 5. With the increasing firing temperature, the values of water absorption do decrease, but their steady state shift due to a more difficult access of water into the pore structure of a brick body.

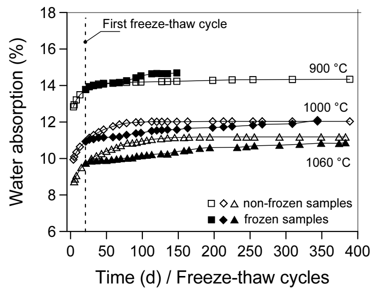

Fig. 5. Effect of water storage and freeze/thaw cycles on the water absorption

When comparing the water absorption values between samples stored only in water (W) and samples exposed to freeze/thaw cycles (F/T) we can see that at the firing temperature of 900 °C their values are virtually identical up to 75 days of storage in water. Then, water absorption values for the samples F/T start to exceed the water absorption values of the samples W until their breach. The water absorption values of samples subjected to freeze/thaw cycles (F/T) are lower on a long-term basis with increasing firing temperature as compared with samples (W) and only very slowly approach these values (e. g. at the temperature of 1000 °C it is after nearly 350 days of storage in water).

Water absorption of a brick body during the freeze/thaw cycles is influenced not only by the gradual change in pore structure, but also by the change of the irreversible expansion. The effect of change in pore volume and change of the median pore radius on water absorption is virtually identical and in both cases, it is a linear dependence, see Figures 6 and 7. In case of the irreversible expansion, it is a continuous trend, where the increase of expansion couples with the increase of absorption, see Fig. 8.

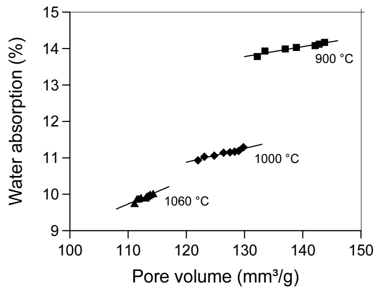

Fig. 6. Relationship between the water absorption and pore volume during freeze/thaw cycles

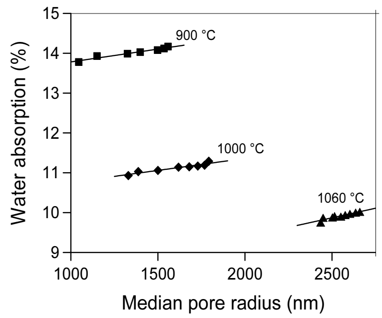

Fig. 7. Relationship between the water absorption and median pore radius during freeze/thaw cycles

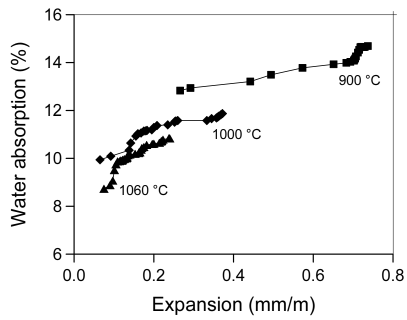

Fig. 8. Relationship between the water absorption and irreversible expansion

Effects of freeze/thaw cycles on the change of pore structure are shown in Figures 9 and 10. The course of changes in pore volumes according the number of freeze-thaw cycles is shown in Figure 9. For all three firing temperatures we follow a continuous process. Volume changes of pores takes place the fastest on the brick body, which was fired at temperature 900 °C. With the increasing firing temperature is the rate of this change slowed down significantly. With a similar trend we can also meet at monitoring of the median pore radius, see Figure 10. The difference between these two properties lies in the fact that with the increase firing temperature the initial values of the pore volume decrease and, on the contrary, the initial value of median pore radius grows. These results confirm the fact that in order to achieve a high frost resistance of brick body, two important conditions have to be met: the pore structure should have the greatest value of the median pore radius (primary condition) and the lowest value of the pore volume (secondary condition) [22, 32].

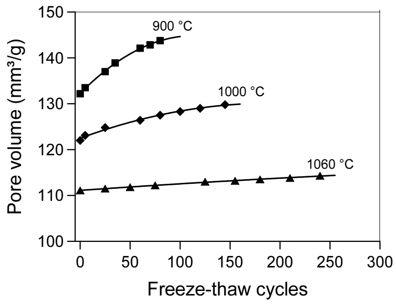

Fig. 9. Dependence of pore volume from the number of freeze/thaw cycles

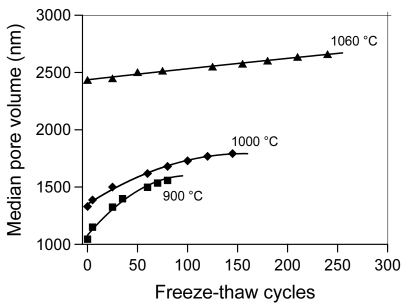

Fig. 10. Dependence of median pore volume from the number of freeze/thaw cycles

More detailed transformations of pore structure during the freeze-thaw cycles are shown in Figures 11 – 13. In these cases, the histograms show the percentage changes in volumes of pores on the brick body at firing temperatures of 900, 1000 and 1060 °C. Based on their increasing or decreasing trend a virtual boundary has been established at the temperature of 900 °C for the pore diameter of 1 µm, at temperature of 1000 °C for the pore diameter of 1.5 µm and at temperature of 1060 °C for the pore diameter of 3.0 µm. From this virtual boundary the volume of small pores gradually decreases with the increase in number of freeze/thaw cycles. This trend can be observed especially at the firing temperature of 900 °C, see Figure 11. With an increasing firing temperature, the trend is no longer so clear.

These changes in the field of pore volume have caused an irreversible change – the increase of values of the median pore radius, see Figure 10.

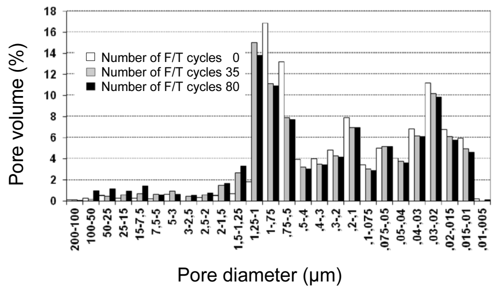

Fig. 11. Change of pore volume according to the diameter and number of freeze/thaw cycles at firing temperature of 900 °C

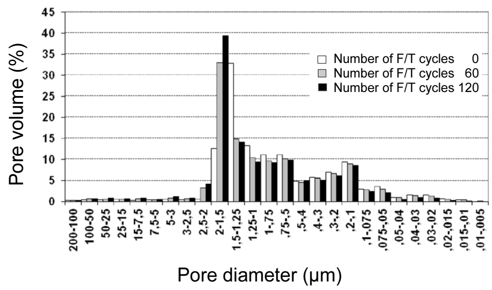

Fig. 12. Change of pore volume according to the diameter and number of freeze/thaw cycles at firing temperature of 1000 °C

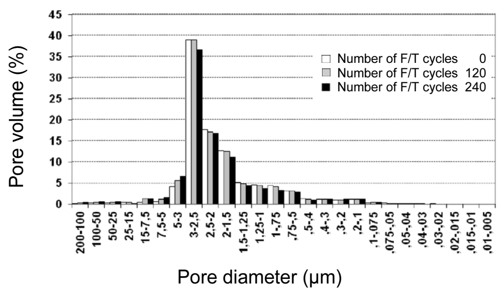

Fig. 13. Change of pore volume according to the diameter and number of freeze/thaw cycles at firing temperature of 1060 °C

5. CONCLUSIONS

Irreversible expansion and water absorption of the brick body in the process of freezing and thawing depend from the firing temperature; these phenomena significantly decrease with the rise of temperature.

The results obtained in this work show that in order to achieve a high frost resistance of brick body, two important conditions have to be met: the pore structure should have the greatest value of the median pore radius (primary condition) and the lowest value of the pore volume (secondary condition).

The change of the porous system during freeze-thaw cycles takes place so that the volume of small pores gradually decreases and conversely, a large pore volume gradually increases. This results in a gradual increase in the value of the median pore radius. In this case, we can see that brick body has a tendency to adjust its pore structure so as to best resist the effects of freeze-thaw cycles.

Interesting results were obtained in the comparison of water absorption values of the samples stored only in water (W) and samples exposed to freeze-thaw cycles (F/T) after 21 days of common storage. Higher water absorption of samples F/T in contrast to the samples W underlines the fact that in case of the F/T samples there have been significant changes in the porous system. This should be a signal that in a short time a total destruction of the brick body will occur.

The knowledge gained can be applied when predicting the durability of older clay roofing tiles or facing bricks, which have been placed in the outdoors for long periods of time. If this product will be subjected to e. g. 50 to 100 freeze-thaw cycles and the values of the pore volume and of the median pore radius will remain practically unchanged, then we can state that the product has a suitable pore structure and can achieve high durability. Conversely, if the values of the pore volume and of the median pore radius show great changes, we can assume that the brick product will no longer performs its function in the short term.

REFERENCES

1. **Setzer, M. J.** Action of Frost and Deicing Chemicals. Basic Phenomena and Testing. In: Marchand, J., Pigeon, M., Zetzer, M. (Eds.) Freeze-thaw Durability of Concrete, E&FN Spon, London 1997: pp. 3 – 21.
2. **Setzer, M. J.** Micro-ice-lens Formation in Porous Solid *Journal of Colloid and Interface Science* 243 2001: pp. 193 – 201.
3. **Chaplin, M.** Water Structure and Science. Water Anomalies *In:* http://www.lsbu.ac.uk/water/anmlies.html.
4. **Ražnjević, K.** Thermodynamic Tables. Publishing House ALFA, 1984, Bratislava.
5. **Seaverson, E. J., Brosman, D. A.** Expansion Phenomena during Freezing of Saturated Bricks and Implications on Frost Resistance *Ziegelindustrie International* 54 (4) 2001: pp. 13 – 19.
6. **Sadunas, A., Bure, D.** Water Migration Processes in Heavy Clay Ceramics under Cyclic Freezing-thawing *Industrial Ceramics* 20 (3) 2000: pp. 153 – 159.
7. **Franke, L., Bentrup, H.** Evaluation of the Frost Resistance of Bricks in Regard to Long Service Life *Ziegelindustrie International* Part 1. 46 (7 – 8) 1993: pp. 483–492, Part 2. 46 (9) 1993: pp. 528 – 536.
8. **Mačiulaitis, R., Kičaitė, A.** Peculiarities of Destruction Mechanism in Ceramic Products under Simulated Exploitation Conditions *Materials Science* (*Medžiagotyra*) 12 (4) 2006: pp. 341 – 345
9. **Wardeh, G., Perrin, B.** Analysis of Strains in Baked Clay Based Materials during Freezing and Thawing Cycles *Journal of Building Physics* 29 (3) 2006: pp. 202 – 217.
10. **Robinson, G. C.** Reversibility of Moisture Expansion *Bulletin of the American Ceramic Society* 64 (5) 1985: pp. 712 – 715
11. **Hauck, D., Hilker, E., Hesle, E.** Moisture Expansion Behavior of Bricks *ZI-Annual, Annual for the Brick and Tile Structural Ceramics* 1989: pp. 47 – 57.
12. **Hanykýř, V., Maryška, M., Bouška, P., Pume, D.** Formation of Fired Ceramic Body and Its Ageing. Part 1. *Silika* 13 (5 – 6) 2003: pp. 130-135; Part 2. 13 (7 – 8) 2003: pp. 187 – 191 (in Czech).
13. **Hanykýř, V., Kloužková, A, Bouška, P., Vokáč, M.** Moisture Expansion of Porous Structural Ceramics *12th International Scientific Conference* April 20 – 22, 2009, Brno, Czech Republic (in Czech).
14. **Sadunas, A., Matschjulaitis, R., Kitscheite, A.** Methodological Problems in Determining the Structural Properties of Heavy Clay Products with High Frost Resistance *Ziegelindustrie International* 44 (7) 1991: pp. 361 – 363.
15. **Friese, P.** Predictions of the Frost Resistance of Bricks *Ziegelindustrie International* 48 (12) 1995: pp. 952 – 963.
16. **Nakamura, M.** Indirect Evaluation of Frost Susceptibility of Building Materials *Bulletin of the American Ceramic Society* 67 1988: pp. 1664 – 1965.
17. **Kičaitė, A., Mačiulaitis, R.** The Service Frost Resistance Aspect of Water Absorption Kinetics in Ceramic Products *Journal of Civil Engineering and Management* 8 2002: pp. 126 – 131.
18. **Freyburg, S., Finger, F. A.** Microstructure Formation and Durability of Bricks *Ziegelindustrie International* 54 (5) 2001: pp. 32 – 41.
19. **Hansen, W., Kung, J. H.** Pore Structure and Frost Durability of Clay Bricks *Materials and Structures* 21 1988: pp. 443 – 447.
20. **Freyburg, S.** Study of the Evaluation of the Durability of Historic Clay Roofing Tiles *Ziegelindustrie International* 49 (2) 1996: pp. 88 – 98.
21. **Elert, K., Cultrone. G., Navarro, C. R., Pardo, E. S.** Durability of Brick Used in the Conservation of Historic Buildings – Influence of Composition and Microstructure *Journal of Cultural Heritage* 4 2003: pp. 91 – 99.
22. **Šveda, M.** Effect of Water Absorption on Frost Resistance of Clay Roofing Tiles *British Ceramic Transactions* 102 (1) 2003: pp. 43 – 44.
23. **Šveda, M., Unčík, S.** Influence of Pore Structure on the Frost Resistance of Brick Products *Ziegelindustrie International* 52 (7) 1999: pp. 80 – 85.
24. **Schmidt, H., Piltz, G.** Untersuchung des Einflusses von mineralischen Zusatzstoffen auf das Porengefüge und die Frostwiderstandsfähigkeit von Verblendziegeln *Sprechsaal* 110 1977: pp. 2 – 15.
25. **Freyburg, S., Schwarz, A.** Influence of the Clay Type on the Pore Structure of Structural Ceramics *Journal of the European Ceramic Society* 27 (2 – 3) 2006: pp. 1727 – 1733.
26. **Cultrone, G., Sebastián, E., Elert, K., de la Torre, M. J., Gazalla, O., Rodriguez-Navarro, C.** Influence of Mineralogy and Firing Temperature on the Porosity of Bricks *Journal of the European Ceramic Society* 24 (3) 2004: pp. 547 – 564.
27. **Sánchez de Rojas, M. I., Marín, F. P., Frías, M., Valenzuela, E., Rodríguez, O.** Influence of Freezing Test Methods, Composition and Microstructure on Frost Durability Easement of Clay Roofing Tiles *Construction and Building Materials* 25 (6) 2011: pp. 2888 – 2897.

28. **Sadunas, A. S.** Influence of the Granulometric Composition on the Frost Resistance of Heavy Clay Ceramics *Keramische Zeitschrift* 45 (3) 1993: pp. 142 – 146.
29. **Šveda, M.** The Effect of Firing Temperature and Dwell Time on the Frost Resistance of a Clay Roofing Tile *Ziegelindustrie International* 57 (6) 2004: pp. 36 – 43.
30. **Ranogajec, J., Kojić, P., Rudić, O., Ducman, V., Radeka, M.** Frost Action Mechanisms of Clay Roofing Tiles: Case Study *Journal of Materials in Civil Engineering* 24 (9) 2012: pp. 1254 – 1260. http://dx.doi.org/10.1061/(ASCE)MT.1943-5533.0000500
31. **Sokolář, R.** Increasing of Durability of Facing Bricks *In: 4 International Conference Building Materials and Testing* 2003, ORGWARE, Štrbské Pleso Slovakia, 2003: pp. 36 – 39 (in Czech).
32. **Šveda, M.** The Effect of Antika Admixture on the Frost Resistance of Clay Roofing Tiles *Ziegelindustrie Int.* 55 (10) 2002: pp. 29 – 33.
33. **Ikeda, K., Kim, H., Kaizu, K., Higashi, A.** Influence of Firing Temperature on Frost Resistance of Roofing Tiles *Journal of the European Ceramic Society* 24 (14) 2004: pp. 3671 – 3677.
34. **Šveda, M., Unčík, S.** The Effect of the Pore Structure of a Brick Body on Black Core Formation in Roofing Tiles Manual 2008 / Tile & Brick International 30 – 35.
35. **Pytlík, P.** Clay Roofing Tile *Silika* 11 (5 – 6) 2001: pp. 158 – 164 (in Czech).
36. **Maage, M.** Frost Resistance and Pore Size Distribution in Bricks. Part 1. *Ziegelindustrie International* 43 (9) 1990: pp. 472 – 481; Part 2 10): pp. 582 – 588.
37. **Arnott, M.** Investigation of Freeze-thaw Durability In: NRC-IRC Report N. CR 5680.1, Nat. Res. Council of Canada, Ottawa, Canada, 1990.
38. **Koroth, R., Fazio, P., Fedman, D.** Development of New Durability Index for Clay Bricks *Journal of Civil Engineering and Architecture* 9 (3) 1988: pp. 26 – 33 and 9 (3) 1998: pp. 87 – 93.
39. **Robinson, G. C.** Relation between Physical Properties and Durability of Commercial Marketed Bricks *Bulletin of the American Ceramic Society* 56 (12) 1995: pp. 1071 – 1075.
40. **Vincenzini, P.** Le prove di laboratorio nella previsione del comportamento al gelo dei materiali ceramici per l'edilizia *Ceramurgia* 3 (4) 1974: pp. 176 – 188.
41. **Raimondo, M., Ceroni, C., Dondi, M., Guarini, G., Marsigli, M., Venturi, I., Zanelli, C.** Durability of Clay Roofing Tiles: the Influence of Microstructural and Compositinal Variables *Journal of the European Ceramic Society* 29 (15) 2009: pp. 3121 – 3128.
42. **Freyburg, S., Schwarz, A.** Influence of the Clay Type on the Pore Structure of Structural Ceramics *Journal of the European Ceramic Society* 24 (2 – 3) 2006: pp. 1727 – 1733.
43. **Wardeh, G., Perrin, B., Laurent, G., Poeydemenge, F.** Freeze-thaw Cycling Resistance of Clay Roofing Tiles *Ziegelindustrie International* 59 (3) 2006: pp. 21 – 30.