BRICKTECH: ASSESSMENT FOR THE USE OF WASTE IN THE BRICK PRODUCTION AND OPTIMIZATION OF THE FIRING CONDITIONS

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Abstract

For millennia clay bricks have been used as building materials for their excellent properties. From both environmental and economic perspectives they are still a valuable, healthy and efficient construction material. Industrial brick production actually is investing in quality and development of new mixes using wastes to save on the extraction of the raw materials. BRICKTECH is a research study based on the knowledge of brick and on its development, considering several aspects: the mineralogical and textural proprieties as a “pyrometamorphic rock”, in particular concerning the evolution of new phases during firing; the porosity, the main features that rule the durability and the physical proprieties of these construction materials; and, finally, the assessment for the use of production waste from natural stone industry such as that of trachyte, reducing environmental impact of building material and creating new mix designs which should be of interest for industry. This research, therefore, contributes to perpetuate the use of a traditional construction material creating new assets for the future generations.

Introduction

Brick can be considered as an artificial metamorphic rock. The raw materials composition, principally constituted by clay minerals, and the firing temperatures strictly control the characteristics of the fired products. During the firing process new minerals grow replacing mineral phases present on micro-domains of reactions, in particular along the grain boundaries, obtaining a polymineral aggregate, with a microstructure and a mineralogy that resembles a “pyrometamorphic rock” (Duminuco et al., 1998; Riccardi et al., 2009).

Another important aspect of bricks is their porosity, especially concerning the pore-size distribution and the pore interconnection. The pore system is the principal parameter to evaluate the durability of bricks, because it connects the building material with the environment and determines the capacity of fluid storage and circulation (Cultrone et al., 2004).

Not least, the theme of waste reuse as raw materials is actually an important requirement of development for industrial brick production for saving resources, energy and costs, and to benefit the environment. The main aspects in which this study is addressed are:
- firing experiments at different temperatures of “ad hoc” prepared mixtures;
- mineralogical, physical and aesthetic characterization of commercial bricks;
- study of the porosity;
- reuse of trachyte as a waste material.

Materials

Firing experiments at different temperatures of “ad hoc” prepared mixtures

Two mixtures (M3 and M4) with different composition were prepared using pure phases and standard clays (Illite IMt-1 – Cambrian shale and Ripidolite CCa-2). Sample M3 was obtained mixing the following constituents: quartz 30 wt.%, calcite 25 wt.%, K-feldspar 5 wt.%, illite 24 wt.%; ripidolite 16 wt.%; whereas sample M4: quartz 28 wt.% , calcite 23 wt.%, K-feldspar 5 wt.%, illite 23 wt.%, ripidolite 15 wt.%, coal-burned 6 wt.%. Clay minerals were < 40 μm in size, while temper grain-size was comprised in the range 63 - 125 μm.

Mineralogical, physical and aesthetic characterization of commercial bricks

Five industrial bricks (called GP, N, R6, RSS and RS), which differ in siliceous and carbonatic contents, were characterized. GP and N were fired at 1050°C, RS at 980 °C, RSS at 950 °C and R6 at 600°C.
Study of porosity
Four of the five industrial bricks (GP, N, R6 and RSS) were chosen to study the distribution and geometry of the pore system. R6 and RSS have the same mineralogical composition, but different firing temperature (600°C and 950 °C, respectively); GP and N are fired at the same temperature (1050 °C), but they are different in composition because N contains Mn-oxides added as dye additive.

Reuse of trachyte as waste material
Three different types of bricks were produced by adding 5, 10 and 15 (% wt.) of trachyte to a clay material collected from the Veneto region, and were fired at 950, 1000 and 1100 °C (samples: MT-5.90, MT-5.10, MT-5.11, MT-10.9, MT-10.10, MT-10.11, MT-15.9, MT-15.10, MT-15.11).

Methods
Texture, mineralogy, chemistry, extent of vitrification and porosity were studied by Optical Petrographic Microscopy (OPM, UNI EN 11084), Scanning Electron Microscopy (SEM), Field Emission Scanning Electron Microscopy (FESEM), X-ray Fluorescence (XRF) and X-ray Powder Diffraction (XRPD). Colorimetry was carried out on wet and dried samples using a Konica Minolta CM-700d spectrophotometer. The hydric parameters were performed using cube-shaped samples (50 mm-edge) (three samples per type of brick) and prismatic-shaped samples (25 x 25 x 120 mm). Water absorption and drying were determined according to the method described in UNI EN 772-7 (1998) normative. Drying index (Di), apparent and real density, open porosity and the degree of pore interconnection were calculated.

Accelerated ageing tests were carried out on 30 cube-shaped samples (50 mm-edge) to evaluate their resistance to frost and salt crystallization as follows: 30 cycles of 24 h duration were performed for freeze-thaw test (UNI EN 12371) and 10 cycles of 24 h duration for salt crystallization test (UNI EN 12370). Ultrasound velocity was measured to check the elastic-mechanical characteristics and structural anisotropy of the fired bricks and to detect variation in the degree of compactness during and after the ageing treatments. Pore size and pore distribution in the range 0.001–100 µm were determined using Mercury Intrusion Porosimetry (MIP). The study of the pore system was completed by Digital Image Analysis (DIA) of back-scattered images acquired by Scanning Electron Microscopy (SEM) and 3D models obtained from Computed micro-Tomography (µ-TC). In order to evaluate bricks thermal insulation, Infrared Thermography (IRT) was performed on cubic samples of 50 mm-edge, using a FLIR T440 series camera. This test consisted in heating the sample from one surface (their base) during 20 min at 50 °C and recording infrared images at regular intervals of 30 seconds.

Results and following planned investigations
Firing experiments at increasing temperature of “ad hoc” prepared mixtures
Firing experiments were carried out by increasing the temperature, with steps of 50°C, from 550 to 1150°C.
XRPD analysis of fired samples showed the progressive decomposition of some phases and the development of new silicate assemblages.
At 750°C the peak intensity of illite decreased and half-peak width widened, while epidolite totally disappeared. At 800 °C the peak of calcite considerably reduced while small peaks of new silicate phases appeared (gehlenite and wollastonite). Starting from 950°C illite totally disappeared and new silicate phases such as sandine formed.

Further planned analyses: mineralogical and textural study with SEM-EDS analysis; FT-IR analysis of gases released during the heating of samples; quantitative analysis of phases based on XRPD data using the Rietveld method.

Mineralogical, physical and aesthetic characterization of commercial bricks
Mineralogical analysis (FESEM-EDS and XRPD) showed that RSS, RS, GP and N samples (fired over 900°C) are characterized by a similar mineralogy (quartz, wollastonite, gehlenite, plagioclase, K-feldspar and diopside), while R6 (fired at 600°C) still contains phyllosilicates, calcite and dolomite.
The highest ultrasonic wave velocities were measured in samples GP and N, while the lowest in sample R6. This suggests that GP and N are the most compact bricks, while R6 is the most porous one. With the increase of firing temperatures, samples became texturally more homogeneous and anisotropy decreases. Poisson ratio (ν) showed similar values for all samples, with a small rising in those with a higher content in carbonates. GP stands out for the highest shear (G) and Young moduli (E).

Data from freeze-thaw and salt crystallization tests showed a similar initial behavior for all samples, except for R6 that resulted to be the weakest. Infrared Thermography (IRT) revealed a qualitative different heating. GP appears the sample more susceptible and shows the fastest heating, while N and R6 are the most refractory.

Further planned analyses: uniaxial compression test.

Pore system
The study of porosity was performed by using different techniques, “direct” and “indirect”, to draft a general characterization of the pore system and overcome their differences and limitations. Mercury Intrusion Porosimetry and Hydric test are considered “indirect” methods because they are based on the movement of fluids inside the material and evaluated only interconnected pores (open porosity). On the other hand, Digital Image Analysis and Computed micro-Tomography are considered “direct” techniques because they permit the observation of shape of pores and, furthermore, to measure the total (closed plus open) porosity. Another important difference to consider is the size of pores that each technique is able to investigate (fig. 1). The different approaches only overlap in a specific interval of pore size. In this specific case, the “overlapping zone” has been identified according to the image resolution established for Digital Image and micro-Tomography analyses.

Digital Image Analysis in 2D was processed using BSE images taken by Scanning Electron Microscopy. In particular this study was performed merging 30 images at magnification of 50x and obtaining a larger representative image for each sample. The overall image was segmented in a binary mode (supervised segmentation method using Multispec® software) and analysed with ImageJ® software to obtain a value of total porosity (%). By separately processing the 30 images and comparing them with the overall image the results highlight the high variability of each image (see standard deviation values in Table 1), and thereby, the low representativeness of a single image covering a small portion of the sample. In particular, it was verified that the presence of large pores in a single image overestimated the value of total porosity. In general, traditional methods (MIP and Hydric tests) provide similar values of (open) porosity, while the results of the other techniques are contradictory. These discrepancies are more evident if we considered only the values inside the “overlapping zone” (1-100 µm).
The present results (Table 1) show the high complexity of this topic and the necessity to refine the approach to be adopted with Digital Image Analysis and micro-Tomography, in particular concerning the resolution and the evaluation of pore size in two and three dimensions.

<table>
<thead>
<tr>
<th>Sample</th>
<th>%tot&gt;10μm</th>
<th>Ømax</th>
<th>%tot&gt;10μm</th>
<th>Ømax</th>
<th>Dev.st.</th>
<th>%min</th>
<th>%max</th>
<th>%mean</th>
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<tr>
<td>R6</td>
<td>5.7</td>
<td>880</td>
<td>17.09</td>
<td>2800</td>
<td>1.91</td>
<td>1.51</td>
<td>8.6</td>
<td>4.5</td>
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<td>RSS</td>
<td>8.83</td>
<td>1830</td>
<td>31.74</td>
<td>400</td>
<td>2.27</td>
<td>4.57</td>
<td>14.9</td>
<td>7.1</td>
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<td>GP</td>
<td>11.5</td>
<td>2710</td>
<td>20.57</td>
<td>90</td>
<td>7.72</td>
<td>4.12</td>
<td>35.4</td>
<td>11.7</td>
</tr>
<tr>
<td>N</td>
<td>-</td>
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</table>

Table 1: Data of pore system study. 2D DIA analyses (SEM-BSE) Panoramic image: %tot>10μm, total porosity of panoramic image with threshold of 5 pixels=10μm; Ømax, maximum value of pore diameter (Feret diameter); 3D analyses-Computed Tomography: %tot>10μm, total porosity of panoramic image with threshold of 2 pixels=10μm; Ømax, maximum value of diameter (Sauter diameter) of pore; 2D DIA Values of single images: Dev. St., Standard Deviation of all total porosity of each image. %min, lowest total porosity value, %max, highest total porosity value, %mean, medium value of all images; MIP, %tot, open porosity value evaluated by Mercury Intrusion Porosimetry; Hydric test: %tot open porosity value estimated with Hydric test.

Further investigations required: the study should be completed for all samples also using additional techniques (nitrogen adsorption test) in order to understand the reasons of these apparent discrepancy and discover a possible way to combine the different analytical methods.

**Reuse of trachyte as waste material**

Colorimetry test highlighted how different contents of trachyte and different firing temperatures modify the aesthetic appearance. This is more evident in dry samples which showed a decrease of the a* and b* parameters with increasing temperature (MT-5.9: a*=15.04, b*=24.38; MT-5.11:a*=13.348, b*=22.958, MT-10.9: a*=14.44, b*=23.24; MT-10.11: a*=12.957, b*=22.4275; MT-15.9: a*=15.48, b*=25.138; MT-15.11: a*=12.73, b*=22.21). XRPD technique identified quartz, plagioclase, K-feldspar, biotite, hematite and diopside. Hydric test showed a rather poor pore interconnection (A<sub>p</sub>). This value is comprised between 9.90 and 15.52, and increases with the increase of firing temperature and trachyte content (Table 2). Drying and capillarity rise are similar for all sample (Di = 0.40; Ks = 0.011-0.016). The values of open porosity determined by hydric test are comprised between 35 and 38%, while sample MT-5.10 has the highest value (50%). The results of open porosity obtained by Mercury Intrusion Porosimetry showed a higher variability, with values comprised between 19 and 38% (Table 2). Ultrasounds test showed good physical-mechanical proprieties and high anisotropy (Table 2). Ageing tests (salt crystallization and freeze-thaw) highlighted a good durability of bricks under stressed environmental conditions, without the presence of damages until the latest cycles of the tests.

Infrared Thermography (IRT) test evidenced a general tendency to heat up quickly (after 20 minutes all samples are half saturated).

<table>
<thead>
<tr>
<th>MT-5.9</th>
<th>MT-5.10</th>
<th>MT-5.11</th>
<th>MT-10.9</th>
<th>MT-10.10</th>
<th>MT-10.11</th>
<th>MT-15.9</th>
<th>MT-15.10</th>
<th>MT-15.11</th>
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<tr>
<td>A&lt;sub&gt;p&lt;/sub&gt;</td>
<td>9.90</td>
<td>10.73</td>
<td>12.12</td>
<td>11.54</td>
<td>12.34</td>
<td>15.52</td>
<td>10.97</td>
<td>11.53</td>
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<tr>
<td>Di</td>
<td>0.40</td>
<td>0.39</td>
<td>0.40</td>
<td>0.40</td>
<td>0.39</td>
<td>0.39</td>
<td>0.40</td>
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<tr>
<td>Ks</td>
<td>0.015</td>
<td>0.016</td>
<td>0.016</td>
<td>0.011</td>
<td>0.016</td>
<td>0.011</td>
<td>0.014</td>
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<tr>
<td>P&lt;sub&gt;o&lt;/sub&gt;</td>
<td>38</td>
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<td>27</td>
<td>38</td>
<td>19</td>
<td>23</td>
<td>36</td>
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<tr>
<td>ΔM</td>
<td>5.09</td>
<td>17.11</td>
<td>13.73</td>
<td>15.42</td>
<td>13.51</td>
<td>8.60</td>
<td>5.49</td>
<td>19.39</td>
</tr>
</tbody>
</table>

Table 2. Hydric parameters: A<sub>p</sub>, degree of pore interconnection (%); Di, drying index; Ks, capillarity coefficient, (g/m<sup>2</sup> s<sup>1/2</sup>); P<sub>o</sub>, open porosity (%). Mercury Intrusion Porosimetry: P<sub>o</sub>, open porosity (%). Ultrasounds: ΔM, total anisotropy (%).

Further planned analyses: characterization of the raw materials and of the mineralogy and texture of the fired bricks with Scanning Electron Microscopy (SEM-EDS); uniaxial compression test on fired samples.
Future plans
- Study equilibrium thermodynamics at a microdomain scale during nucleation and growth of new mineral phases (EBSD analysis with FESEM);
- Experimental firing with the increase of temperature (from 550 to 1150 °C, with steps of 50 °C) on three different semi-industrial types of bricks prepared with the addition of trachyte (5, 10 and 15 % in wt.); study of textural and mineralogical evolution with temperature (XRPD, SEM-EDS, FT-IR);

References

SUMMARY OF ACTIVITY IN THIS YEAR
Courses:
- GODARD G., MASSIRONI M., PERUZZO L.: Acquisizione, trattamento e analisi di micro-immagini: applicazioni ai materiali naturali ed artificiali, 2-3 April 2014, Padova, Italy
- 3rd International Workshop on Mortar Dating, 14-16 April 2014, Padova, Italy

Congresses:
- COLETTI C., CULTRONE G., MARITAN L., MAZZOLI C.: Combined multianalytical approach for the characterization of commercial bricks with a view to their technical use. XXXIV Reunión de la SEM (Sociedad Española de Mineralogía), 1-4 July 2014, Granada, Spain
- COLETTI C., MARITAN L., MAZZOLI C., CULTRONE G.: Characterization of mineralogy and porosity of commercial bricks: 2D and 3D geometry analysis of the pore system. SGI-SIMP 2014, 10-12 September 2014, Milano, Italy

Publication:
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