Predicting The Initial Rate of Water Absorption in Clay Bricks

Mariarosa Raimondo
Michele Dondi
Davide Gardini
Guia Guarini

ABSTRACT

In this work the capillary suction behaviour of fifteen clay bricks was investigated (UNI 10859) in order to assess the effect of product characteristics and processing conditions and to verify the reliability of the available models. The suction behaviour of bricks, in terms of both total amount of liquid absorbed and kinetics of the process, can basically be brought back to the models of Gummerson et co-workers and Hoffman and Niesel; these latter, however, do not make provision for the effect of product microstructure and water physical properties on the liquid-porous solid interactions. Even if experimentally the liquid suction presents a first dependence on the amount of porosity - a less dense ceramic body is undoubtedly able to absorb a larger liquid amount - other characteristics of the porous structure (size, tortuosity and internal specific surface of the capillary system, as well as the presence of mineralogical phases) request to be more deeply investigated since they could affect the process kinetics.

According with the Beltran’s model, which revealed to be sufficiently reliable, the values of capillary coefficient $K_s$ were calculated and their correlation with the experimental ones has been provided. The results obtained indicate that, for a given liquid and in the same experimental conditions, varying in a controlled way the product microstructure (i.e. decreasing the pore size, increasing the pore tortuosity and/or controlling the amorphous/new formed phases ratio) should allow to design materials having a more suitable behaviour.

KEYWORDS

Clay brick, Capillary absorption, Porosity, Microstructure, Phase composition

1  Institute of Science and Technology for Ceramics, ISTEC-CNR, Faenza, Italy 48018, Phone +39 0546 699718, Fax 0546 46381, mrosa@istec.cnr.it
2  Institute of Science and Technology for Ceramics, ISTEC-CNR, Faenza, Italy 48018, Phone +39 0546 699728, Fax 0546 46381, dondi@istec.cnr.it
3  Institute of Science and Technology for Ceramics, ISTEC-CNR, Faenza, Italy 48018, Phone +39 0546 699749, Fax 0546 46381, davide@istec.cnr.it
4  Institute of Science and Technology for Ceramics, ISTEC-CNR, Faenza, Italy 48018, Phone +39 0546 699718, Fax 0546 46381, guiagrn@istec.cnr.it
1 INTRODUCTION

In the structural design of masonry, the suction behaviour of clay bricks has to be strictly controlled in order to optimize the mechanical performances and to prevent deterioration processes. Clay bricks capacity of absorbing liquids and moisture, from both the immediate surrounding and the other building materials, can be referred as their “capillarity”. Since clay bricks have to satisfy many requirements (i.e. thermal and acoustic properties, load-bearing capacity and ecological impact), the relationship between these properties and capillarity phenomenon goes through their microstructural characteristics and, in particular, amount, size and shape of pores.

In the literature, a time dependent law of fluid absorption by porous materials, taking the form:

\[ m = K_s \cdot t^{1/2} \]  

(1)

has been proposed [Gummerson et al., [1981]]; the water absorbed by a porous solid per surface unit area \((m)\) increases as the square root of the elapsed time \((t)\), with \(K_s\) being defined as the capillary coefficient. In the last years, other mathematical functions were empirically derived [Hoffman et al. [1985, 1990]] and the simple relation (1) was replaced by an exponential one:

\[ m = a \cdot [1-\exp(-bt^{1/2})] \]  

(2)

where \(a\) is the maximum moisture content and \(b\) a factor influencing the suction kinetics; multiplying \(a\) per \(b\), the liquid absorption coefficient is obtained. Equation (1) was furtherly developed [Beltrán et al. [1988]] considering that the capillary coefficient \(K_s\) must depend on both properties of liquid and of porous solid. Accordingly, the capillary coefficient can be expressed by the formula:

\[ K_s = \frac{\rho \gamma}{\mu} \cdot \left( \frac{OP}{\lambda} \right)^{1/2} \cdot r_0^{1/2} \cdot \left[ \cos \theta / 2 \right]^{1/2} \]  

(3)

where \(\rho\), \(\gamma\) and \(\mu\) are, respectively, the density, the surface tension and the viscosity of the liquid, while \(OP\) represents the open porosity accessible by the liquid, \(r_0\) the median pore size and \(\lambda\) the pore tortuosity of the solid; \(\theta\) is the liquid-brick contact angle.

For a given temperature, grouping the liquid properties and the contact angle term into a constant \(C\), the equation (3) is reduced to:

\[ K_s = C \cdot \left( \frac{OP}{\lambda} \right)^{1/2} \cdot r_0^{1/2} \]  

(4)

while equation (1) can be written in the form:

\[ m = [C \cdot \left( \frac{OP}{\lambda} \right)^{1/2} \cdot r_0^{1/2}] \cdot t^{1/2} \]  

(5)

When the experimental results are represented graphically in the form of \(m\) versus \(t^{1/2}\), a straight line with a slope \(K_s\) should be observed.

The aims of this work are: i) giving a representative survey of the suction behaviour of clay bricks and ii) assessing the influence of the main microstructural and compositional parameters of clay bricks on the absorption process. The results pursued could allow to improve the evaluation of the effects of product and processing conditions on the capillary suction of porous bodies, thus representing an important starting point in the structural design of masonry components.
2 MATERIALS AND METHODS

Fifteen samples of clay bricks were collected in different brickworks, representing the Italian production. Products were characterized by the determination of phase composition, open and total porosity, bulk density, pore size distribution and pore specific surface. Phase composition was quantitatively determined by X-ray powder diffraction (Rigaku Miniflex, CuKα radiation) with the Reference Intensity Ratio method (Al₂O₃ as internal standard, experimental error is within 5% relative). Open porosity (OP) and bulk density (BD) were quantified according to ASTM C373. Specific weight (SW) was measured by He pycnometry (Micromeritics MVP 1305, ASTM C329), while total porosity (TP) was calculated by the equation: TP = (1-BD/SW) · 100.

Pore size distribution (in the 0.01 – 100 μm range) was determined by MIP (Thermo Finnigan Pascal 140/240) with an experimental uncertainty of about 1% relative. Porosimetric data are expressed as r₀ (median pore radius), amount of micropores (P50) having a diameter < 0.05 μm (i.e. < 50 nm) and amount of macropores (P3) having diameter > 3 μm. Pore specific surface (SS) analyses were performed by nitrogen absorption (Micromeritics FlowSorb II 2300) following the B.E.T. single point method.

The capillary absorption of samples was determined according to UNI 10859 on circular disks (20.0 ± 0.1 cm diameter, thickness between 1.5 and 2.0 cm). Samples were dried in an electric oven at 60 ± 2°C for 7 days and, after cooling, their weight (m₀) was measured. A basal face of each disk was put into direct contact with a 1 cm layer of paper, filled with distilled water at 20°C, in a closed vessel. The mass of specimens (mᵢ) was recorded after 10, 20, 30 minutes and 1, 4, 6, 24, 48 and 72 hours and the liquid mass absorbed per surface unit area (m) was calculated by the formula:

\[ m = \frac{(mᵢ - m₀) \cdot A}{1} \]  

where A is the area of the specimen in contact with the paper layer.

The capillary absorption of each sample was described plotting m versus the square root of the elapsed time (Fig.1). The graph obtained presents an initial straight line, whose slope is the experimental capillary coefficient Kₛ, and a final stage characterized by an asymptotic trend.

![Figure 1. Suction behaviour of clay bricks.](image-url)

In order to verify equation (3), the following physical properties of water at 20°C were utilized (3): density \( ρ = 0.998 \text{ g cm}^{-3} \); surface tension \( γ = 72.7 \text{ mN m}^{-1} \); viscosity \( μ = 1.0 \text{ mPa s} \). The capillary tortuosity factor \( λ \), which is an empirical dimensionless parameter, was calculated as the ratio:

\[ λ = \frac{r₀ \cdot SS \cdot BD}{2 \cdot OP} \]  

(7)
The water-brick contact angle was measured on the brick surface (DataPhysics Instrument OCA15) obtaining an average value of $\theta = 87\pm 3^\circ$. A statistical elaboration of data was performed by linear binary correlation and multivariate analysis techniques (factor analysis, scree test for eigenvalues) using the StatSoft Statistica 6.0 software.

3 RESULTS AND DISCUSSION

3.1 Physical Properties

Samples show a great variability of their physical and microstructural parameters [Table 1]: total porosity is in the 19-43 %vol. range, mostly represented by open porosity (19-40 %vol). The median pore size $r_0$ is between 0.1 and 1.0 $\mu$m, while specific surface varies in the 0.6-2.4 $m^2 g^{-1}$ range. An exception is represented by the sample SLA, which has the smallest pores ($r_0 = 0.09 \mu$m) developing a high specific surface ($12.3 m^2 g^{-1}$). The fraction of pores having dimensions under 50 nm (P50) is quite significant in sample S (13.4 %vol), despite being the less porous material. On the other hand, the amount of the larger pores (P3) is maximum in samples RIN, MO and WPP.

<table>
<thead>
<tr>
<th>Samples</th>
<th>OP (%vol)</th>
<th>TP (%vol)</th>
<th>BD (g cm$^{-3}$)</th>
<th>$r_0$ (nm)</th>
<th>SS ($m^2 g^{-1}$)</th>
<th>P50 (%vol)</th>
<th>P3 (%vol)</th>
<th>$\lambda$ (adim.)</th>
<th>Thickness (cm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>AT</td>
<td>39.0</td>
<td>42.6</td>
<td>1.605</td>
<td>0.48</td>
<td>2.40</td>
<td>2.7</td>
<td>1.2</td>
<td>2.4</td>
<td>1.83</td>
</tr>
<tr>
<td>CA</td>
<td>33.3</td>
<td>34.9</td>
<td>1.763</td>
<td>0.66</td>
<td>2.00</td>
<td>0.9</td>
<td>1.1</td>
<td>3.6</td>
<td>1.95</td>
</tr>
<tr>
<td>CAN</td>
<td>36.0</td>
<td>39.3</td>
<td>1.696</td>
<td>0.61</td>
<td>2.02</td>
<td>0.8</td>
<td>0.8</td>
<td>2.9</td>
<td>1.49</td>
</tr>
<tr>
<td>CAP</td>
<td>36.7</td>
<td>38.4</td>
<td>1.684</td>
<td>0.61</td>
<td>1.87</td>
<td>0.1</td>
<td>0.6</td>
<td>2.6</td>
<td>1.65</td>
</tr>
<tr>
<td>F</td>
<td>24.4</td>
<td>27.3</td>
<td>1.870</td>
<td>0.42</td>
<td>1.00</td>
<td>0.6</td>
<td>0.5</td>
<td>1.6</td>
<td>1.55</td>
</tr>
<tr>
<td>ILS</td>
<td>27.7</td>
<td>37.5</td>
<td>1.716</td>
<td>0.38</td>
<td>1.90</td>
<td>1.5</td>
<td>1.1</td>
<td>2.2</td>
<td>1.84</td>
</tr>
<tr>
<td>MO</td>
<td>36.3</td>
<td>36.5</td>
<td>1.766</td>
<td>0.43</td>
<td>1.20</td>
<td>0.7</td>
<td>4.5</td>
<td>1.3</td>
<td>2.02</td>
</tr>
<tr>
<td>RIN</td>
<td>33.3</td>
<td>36.2</td>
<td>1.712</td>
<td>0.99</td>
<td>2.11</td>
<td>4.2</td>
<td>4.8</td>
<td>5.4</td>
<td>1.47</td>
</tr>
<tr>
<td>RIP</td>
<td>32.7</td>
<td>34.0</td>
<td>1.715</td>
<td>1.00</td>
<td>1.47</td>
<td>5.8</td>
<td>4.2</td>
<td>3.9</td>
<td>1.63</td>
</tr>
<tr>
<td>S</td>
<td>18.8</td>
<td>18.8</td>
<td>2.121</td>
<td>0.61</td>
<td>0.60</td>
<td>13.4</td>
<td>1.6</td>
<td>2.1</td>
<td>1.62</td>
</tr>
<tr>
<td>SLA</td>
<td>28.9</td>
<td>28.9</td>
<td>1.896</td>
<td>0.09</td>
<td>12.30</td>
<td>0.8</td>
<td>1.2</td>
<td>3.5</td>
<td>2.02</td>
</tr>
<tr>
<td>WPP</td>
<td>36.5</td>
<td>38.6</td>
<td>1.657</td>
<td>1.00</td>
<td>1.61</td>
<td>3.1</td>
<td>6.1</td>
<td>3.7</td>
<td>2.15</td>
</tr>
<tr>
<td>WSN</td>
<td>35.2</td>
<td>38.1</td>
<td>1.673</td>
<td>0.84</td>
<td>1.40</td>
<td>0.8</td>
<td>2.7</td>
<td>2.8</td>
<td>2.07</td>
</tr>
<tr>
<td>ZAN</td>
<td>38.5</td>
<td>41.0</td>
<td>1.632</td>
<td>0.54</td>
<td>2.30</td>
<td>0.8</td>
<td>1.2</td>
<td>2.6</td>
<td>1.62</td>
</tr>
<tr>
<td>ZAP</td>
<td>38.4</td>
<td>41.4</td>
<td>1.627</td>
<td>0.56</td>
<td>2.36</td>
<td>0.6</td>
<td>2.2</td>
<td>2.8</td>
<td>1.58</td>
</tr>
</tbody>
</table>

As far as phase composition, samples are mainly made up of quartz (17-45%), with residual K-feldspar and plagioclase up to 25 and 17%, respectively. Illite/mica is also present in most samples, with AT and SLA showing a content as high as 20%. The amorphous phase ranges from 6% (samples CAP and ZAP) to 45% (sample S), while the total amount of pyroxene, wollastonite and melilite fluctuates between 2 and 46%.

3.2 Initial Rate of Water Absorption

The suction behaviour of samples is illustrated in Fig.1. Most of them reach saturation conditions after less than 3 hours, showing then an asymptotic trend involving very small variations of the liquid mass absorbed; nevertheless, the total amount of water absorbed varies from 0.05 g cm$^{-2}$ of sample S to about 0.16-0.17 g cm$^{-2}$ of samples AT, MO, WPP and WSN [Table 2]. On the whole, samples seem to follow two main trends; most present an initial water uptake characterized by a steep slope ($K_s$ values
between 0.6 and 5.9 \( \cdot 10^{-3} \) g cm\(^{-2}\) s\(^{-1/2}\), Fig. 1 and Table 2) that suddenly develops towards the asymptote, while samples S, ILS and SLA exhibit a more gradual behaviour (\(K_s\) between 0.6 \( \cdot 10^{-3}\) and 1.5 \( \cdot 10^{-3}\) g cm\(^{-2}\) s\(^{-1/2}\), Fig. 1 and Table 2). These different trends can be brought back, respectively, to the models proposed by Gummerson and co-workers [1981] or Hoffmann and Niesel [1988]. In particular, the resistance (in terms of both \(m\) and \(K_s\)) offered by S, ILS and SLA to capillary suction has a first correspondence with their pore size and amount: the very low porosity and the significant percentage of micropores of sample S, as well as the smaller pores, when compared to the mean pore size of the other products, present in both ILS and SLA, confirm somehow the conclusions of the literature [Latridou and Ozouf [1978] [Vos and Tammes [1969]]).

### 3.3 Capillary Suction vs Microstructural Properties

Since physical parameters of water can be considered constant in the experimental conditions, liquid absorption and \(K_s\) should depend only on the solid characteristics. A positive correlation of both OP (Fig. 2A) and TP (Fig. 2B) with \(K_s\) seems to exist though with a quite low statistical significance (\(r^2 = 0.524\) and 0.347, respectively) mainly due to the sample MO, which presents a “capillary capacity” much higher than expected on the basis of its porosity. Hence, if it is true that a less dense ceramic body is able to absorb a higher liquid amount, for the same porosity the influence of other parameters characterizing the capillary structure is not obvious.

<table>
<thead>
<tr>
<th>Samples</th>
<th>(m) (g cm(^{-2}))</th>
<th>(K_s \cdot 10^3) (g cm(^{-2}) s(^{-1/2}))</th>
<th>(K_s^* \cdot 10^3) (g cm(^{-2}) s(^{-1/2}))</th>
</tr>
</thead>
<tbody>
<tr>
<td>AT</td>
<td>0.164</td>
<td>3.1</td>
<td>15.4</td>
</tr>
<tr>
<td>CA</td>
<td>0.144</td>
<td>3.3</td>
<td>10.4</td>
</tr>
<tr>
<td>CAN</td>
<td>0.124</td>
<td>2.8</td>
<td>13.2</td>
</tr>
<tr>
<td>CAP</td>
<td>0.139</td>
<td>3.0</td>
<td>15.2</td>
</tr>
<tr>
<td>F</td>
<td>0.095</td>
<td>1.9</td>
<td>13.3</td>
</tr>
<tr>
<td>ILS</td>
<td>0.125</td>
<td>1.5</td>
<td>10.8</td>
</tr>
<tr>
<td>MO</td>
<td>0.165</td>
<td>5.9</td>
<td>25.1</td>
</tr>
<tr>
<td>RIN</td>
<td>0.100</td>
<td>2.2</td>
<td>8.4</td>
</tr>
<tr>
<td>RIP</td>
<td>0.108</td>
<td>2.3</td>
<td>11.7</td>
</tr>
<tr>
<td>S</td>
<td>0.052</td>
<td>0.6</td>
<td>9.7</td>
</tr>
<tr>
<td>SLA</td>
<td>0.138</td>
<td>1.5</td>
<td>3.4</td>
</tr>
<tr>
<td>WPP</td>
<td>0.165</td>
<td>3.4</td>
<td>13.4</td>
</tr>
<tr>
<td>WSN</td>
<td>0.155</td>
<td>3.5</td>
<td>15.7</td>
</tr>
<tr>
<td>ZAN</td>
<td>0.121</td>
<td>3.1</td>
<td>15.2</td>
</tr>
<tr>
<td>ZAP</td>
<td>0.139</td>
<td>3.1</td>
<td>13.9</td>
</tr>
</tbody>
</table>

Concerning the relationships of \(K_s\) with \(r_0\) (Fig. 2C) and P50 (Fig. 2D), the following conclusions can be drawn: i) the suction kinetics is not dependent on the pore size (\(r^2 = 0.015\)) since, for most samples, very different pore dimensions correspond to quite similar values of \(K_s\); ii) the correlation of \(K_s\) with P50 gives rise to a cloudy distribution of data (\(r^2 = 0.269\)) also because, among the samples, there are not big differences in the relative amount of micropores; at all events, the behaviour of sample S (P50 as high as 13.4%) suggests that such micropores could represent an effective restriction to the liquid uptake. In addition, when \(K_s\) is plotted against P3, a correlation with low statistical significance is obtained, so that the statement that larger pores should be the most involved in the suction phenomenon [Beltrán et al. [1988]] was proved to be not completely reliable.
3.4 Predicting the Capillary Coefficient $K_s$

The reliability of the model proposed by Beltrán and co-workers [1988-1999] was checked introducing into the equation (3) the physical data of water at 20°C and, for each sample, the experimental values of open porosity, median pore size, water-brick contact angle ($\theta = 87^\circ$) and tortuosity factor as calculated in the present work. The predicted $K_s$ values obtained range from $3.4 \cdot 10^{-3}$ (g cm$^{-2}$ s$^{-1/2}$) of sample SLA to $25.1 \cdot 10^{-3}$ (g cm$^{-2}$ s$^{-1/2}$) of sample MO ($K_s^*$, Table 2) and their correlation with the experimental ones being illustrated in Figs. 3A and B; fitting the results by linear regression, the straight line going through the origin ($r^2 = 0.692$) provides the following relationship: the value of the capillary coefficient $K_s^*$ predicted on the basis of equation (3) has to be multiplied by 0.212 to obtain the experimental one so that the real suction kinetics of all samples is proved to be lower than expected on the basis of model.

![Figure 2](image1.png)

**Figure 2.** Experimental capillary coefficient $K_s$ vs. (A) open porosity, (B) total porosity, (C) mean pore size and (D) amount of pores $< 0.05 \mu$m of clay bricks.

![Figure 3](image2.png)

**Figure 3.** (A) The experimental $K_s$ vs. the predicted $K_s^*$ values of capillarity coefficient and (B) the capillary coefficients vs. the term $OP/\lambda_0$ ($r_0$)$^{1/2}$.

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Focusing our attention on these aspects, both predicted and experimental values of the capillary coefficient were plotted as function of the term $\frac{OP}{\lambda (r_0)^{1/2}}$ (Fig. 3B); the linear fitting provides straight lines having, respectively, the form:

$$K_s = 2.915 \frac{OP}{\lambda} (r_0)^{1/2} \quad (r^2 = 0.692) \quad (8)$$

and

$$K_s^* = 13.760 \frac{OP}{\lambda} (r_0)^{1/2} \quad (r^2 = 1) \quad (9)$$

The ratio between the slope values ($2.915/13.760 = 0.212$) coincides with the slope of the straight line fitting the experimental/predicted $K_s$ relationship. Moreover, the slope value of equation (8) must correspond to the term: $\rho \left( \frac{\gamma}{\mu} \right)^{1/2} \left[ \cos \frac{\theta}{2} \right]^{1/2}$ which, considering constant the physical data of water at 20°C, provides the value of the water-brick contact angle fitting the experimental results: $\theta = 89^\circ$, very similar to the experimental average value ($\theta = 87^\circ$) utilized in the present work.

Looking back at equation (8) and Fig.3B, it is reliable to conclude that, besides some exceptions, the Beltrán’s model is sufficiently satisfied: for the same value of open porosity, decreasing the pore size and/or increasing the pore tortuosity means to limit the suction rate and, hence, the capacity of clay bricks of absorbing moisture. At any event, this also means that, varying in a controlled way the product $OP/\lambda (r_0)^{1/2}$, it could be possible to design clay bricks having a more suitable suction behaviour and, at least, to bound the structural deterioration of masonry optimizing the performance of its components.

3.5 Capillary Suction vs Phase Composition

During firing, the decomposition of CaCO$_3$, with the formation of Ca-silicates by reaction with the clay minerals and release of CO$_2$, promotes a greater amount of porosity; theoretically, this circumstance is expected to cause higher values of $K_s$ as it is somehow confirmed (Fig. 4A). On the other hand, the amount of amorphous phase, together with the low sintering degree achieved at the brick firing temperature (900-1000°C), involve the presence of a smaller porosity, which, in turn, seems to be a restriction to the liquid suction phenomenon (Fig.4B).

![Figure 4. Experimental capillary coefficient $K_s$ vs. amount of (A) Ca-silicates and (B) amorphous phase of clay bricks.](image)

A statistical treatment of data was also undertaken through the extraction of the principal components: in the procedure, the amount of Ca-silicates and amorphous phase, other than $K_s$, were also considered as variables in addition to the microstructural ones ($OP$, $TP$, $r_0$, $P50$, $P3$, $SS$, $\lambda$). The positive or negative influence of each variables on $K_s$ clearly stands out when the factorial weights are plotted grafically (Fig. 5): a higher porosity, in terms of both open and total values, makes the liquid uptake faster. The restrictive role played by the amount of micropores and the amorphous phase on the water absorption is also evident.
suction rate is also underlined by the statistical procedure, as well as the opposite role played by the new formed Ca-silicates.

Figure 5. Principal components analysis: factors extracted.

4 CONCLUSIONS

On the whole, the suction behaviour of clay bricks follows the models of Gummerson et al. [1981] and Hoffmann et al. [1988]; however, they do not provide any information about the liquid-solid interactions, so that no prediction of the material behaviour in working conditions can be done. The suction capacity of bricks, in fact, is expected to depend on their microstructural characteristics, in particular, amount, size and shape of pores. Besides some exceptions, the linear relationships between the capillary coefficient $K_s$ and these microstructural variables substantially confirm the role played by open porosity in increasing the absorption capacities of clay bricks, while the influence of pore characteristics on the suction kinetics does not clearly stand out.

The values of the capillary coefficient $K_s$ were also calculated on the basis of Beltrán model [1988], whose effectiveness in predicting $K_s$ values proved to be quite good. This means that, varying in a controlled way the term $OP/\lambda (r_0)^{1/2}$, it is possible to design materials having a predictable suction behaviour, hence to bound the damages in terms of structural deterioration. The capillary coefficient $K_s$, together with the microstructural variables and phase composition, finally underwent a statistical procedure that confirmed the influence of porosity, as well as of a coarser pore dimension (in terms of both radius and percentage of pores greater than 3 μm) in increasing the liquid adsorbing rate with the highest statistical significance. In addition, the sintering pattern of products, leading to a different amorphous/crystalline phases ratio, proved to be relevant on the definition of the most suitable microstructure: the higher porosity, promoted by the complete CaCO$_3$ decomposition, and the smaller pore size, connected with the low sintering degree of clay bricks, work in the opposite way.

REFERENCES


