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# Equilibrium moisture content of clay bricks: the influence of the porous structure

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Abstract
The comprehension of the influence exerted by the material microstructure on the hygrometric properties of clay bricks plays a fundamental role in order to control the condensation phenomena and to avoid the deterioration of the masonry structure. The equilibrium moisture content (MEq) of ordinary and lightweight clay bricks was measured and the correlation with microstructure and pore morphology was investigated. The influence of the pore size and specific surface on the amount of MEq was found to be prevalent when compared to the other physical variables. A statistical model was also set up in order to predict the MEq values.

*Keywords*: Clay bricks, water vapour absorption, equilibrium moisture content, microstructure, pore size.

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## 1. Introduction

The deterioration of porous building materials is mainly due to the structure/water interactions, which, depending on the material characteristics, the physical state of water (liquid or vapour) and the environmental conditions, may be of different magnitude involving different sorption mechanisms [1]. In particular, the absorption of humidity from air, the capillary rise, the rain penetration and the condensation phenomena can lead to the formation of superficial moisture and, consequently, to the wearing and tearing of the structure [2,3].
The knowledge of the hygrometric properties of clay bricks and the influence exerted by the material microstructure plays a fundamental role in the assessment of these phenomena. Furthermore, the moisture content of clay bricks is of substantial significance for the insulation properties of the masonry, affecting considerably its thermal conductivity [4-8].
Notwithstanding the interest of this subject, few studies in the literature dealt with the water/masonry interactions; most of them concern the liquid capillary suction properties, with determination of the water absorption rate and the suction coefficient [9-14], while just few others focused on the water vapour permeability [15-17] or moisture content at equilibrium [4-8]. Generally, the available studies describe the water (liquid or vapour) transfer through porous materials and the storage of the residual humidity in their structure, using different physical and mathematical models which require the knowledge to some extent of the pore

system, at least in terms of porosity and pore size distribution. However, a detailed microstructural characterization of clay bricks is often lacking, making it difficult any appraisal of the different approaches and models.
The measurement of the moisture content at equilibrium (hereafter MEq) of ordinary and lightweight bricks was already performed [4-6], pointing out some correlations between the MEq on one hand and the pore specific surface, bulk density, total porosity and mean pore size on the other hand. The MEq was found to be positively related to the pore specific surface, but no significant data about the relationships with other physical or textural parameters came out. Anyway, for most authors the bulk density is the only parameter considered to compare the thermo-hygrometric performances of different clay bricks, even if this variable proved to be unreliable to predict the MEq values [5-6].
This study is aimed at the experimental determination of the MEq on a series of ordinary and lightweight clay bricks, in order to draw a picture representative of their hygrometric properties. The problem was approached by the material point of view, in order understand better the affinity of the brick structure towards the humidity absorption. In this standview, the influence of brick microstructure (particularly amount, shape and size of pores) on MEq was thoroughly investigated, through a statistical analysis of data by binary and multivariate techniques, trying to develop a simple predictional model of the MEq.

## 2. Materials and methods

Two different typologies of industrially manufactured clay bricks were collected, as both lightweight (AL and SL) and ordinary products (A and S). For each brick typology, 10 specimens were prepared by cutting 20 x 10 x 8 mm bars out the industrial products, which were then dried at 105 $\pm$ 0.5°C until constant weight. The amount of absorbed water in function of time was monitored over several weeks in a climatic cell at six different relative humidity values (20, 40, 50, 60, 70 and 80%) under isothermal conditions (22°C). Once the asynthotic limit was approached, the MEq was calculated as the amount of absorbed water per dry volume (kg m-3) with an experimental error around 3% relative.
The bricks were characterized by the physical and microstructural points of view. Open porosity (OP) and bulk density (BD) were determined by water saturation and Archimede's principle, by measuring the dry weight, the water-saturated weight and the weight suspended in water, according to ASTM C 373. Total porosity (TP) was calculated as TP = 1 – (BD/SW), where SW is the specific weight measured by water pycnometry, according to ASTM C 329. The experimental uncertainty is as low as 0.2 % vol (OP and TP) and 0.002 kg dm-3 (BD).
The pore size distribution (in the 0.005–100 $\mu$ m range) was determined by mercury intrusion porosimetry (Carlo Erba Porosimeter 2000); the mean pore size (MPS), the percentage of pores in the 10-50 nm range (P50), the kurtosis of the distribution (Kurt) were drawn out with an experimental uncertainty of about 1% relative.
The pore specific surface analysis was performed by nitrogen absorption (Micromeritics FlowSorb II 2300) following the BET single point method according to ASTM C 1069 (analytical error about 5% relative).
A statistical elaboration of data was performed by linear binary correlation and multivariate techniques (factor analysis, multiple linear regression analysis) using the StatSoft Statistica 5.0 software. Factor analysis was carried out on the main physical and microstructural variables extracting principal components (2 factors according to the scree test for eigenvalues). Multiple linear regression was executed by the forward stepwise method, including intercept in the model and setting F = 1.00 to enter and F = 0.00 to remove.

## 3. Results and discussion

### *3.1. Moisture content at equilibrium*

The determination of the MEq values at different relative humidities provided strictly correlated results: the higher the relative humidity, the larger the amount of water absorbed. In the correlation with the ceramic porous structure, only the MEq measured at a relative humidity of 70% has been reported, since it is easily related to the moisture content measured in the other different conditions by the equation shown in figure 1.

The MEq of samples A and $A_L$ is, respectively, in the 4-6 kg $m^{-3}$ and 10-12 kg $m^{-3}$ ranges, while the MEq values shown by sample S (12-15 kg $m^{-3}$) and $S_L$ (5-11 kg $m^{-3}$) follow the opposite trend, being the lightweight brick less hygroscopic (Table 1). The comparison of these data with those found in the literature is difficult mainly for two reasons: the different size of the specimens (slabs [4,5] or ground brick [6]), and the different experimental conditions (relative humidity and temperature).

However, the equilibrium moisture content of granular materials from 20 different walling bricks (2-32 kg $m^{-3}$ at 86% of relative humidity) presented by Schmidt [6] accounts for very large differences in terms of hygroscopic behaviour. This great dispersion of data is attributed by the author to some factors directly affecting the increase in weight, such as the possible formation of $Ca(OH)_2$, caused by the moisture absorption of ceramic bodies containing CaO, or the underfiring of some samples, which brings about the presence of more reactive sheet silicates as well as a larger internal surface.

The equilibrium moisture content of ordinary and lightweight products, fired at two different temperatures (850 and 1050°C), was also measured by Rimpel [7, 8]; the moisture absorbed by underfired samples (25-52 kg $m^{-3}$) is considerably higher than that absorbed by the samples fired at the higher temperature (1.3-6.5 kg $m^{-3}$). Moreover, the lightweight products generally show a lower capacity of moisture absorption.

### *3.2. Relationships with brick microstructure*

The physical and microstructural characterization of clay bricks is reported in table 1.

The amount of open porosity is always higher in samples A (32.0 $\pm$ 0.1%) and $A_L$ (35.2 $\pm$ 0.1%) if compared to samples S (25.3 $\pm$ 0.1%) and $S_L$ (27.5 $\pm$ 0.1%). Although it varies in narrower ranges, total porosity follows the same trend with slightly higher average values presented by samples A (34.7 $\pm$ 0.1%) and $A_L$ (36.2 $\pm$ 0.1%) with respect to samples S (30.0 $\pm$ 0.1%) and $S_L$ (33.1 $\pm$ 0.1%). The difference between TP and OP is always positive and seems to affect negligibly the hygroscopic behaviour of clay bricks.

Bulk density values correspond well to the differences observed in terms of porosity, being 1.773 $\pm$ 0.002 kg $dm^{-3}$ and 1.706 $\pm$ 0.002 kg $dm^{-3}$ for samples A and $A_L$ respectively, and 1.825 $\pm$ 0.002 kg $dm^{-3}$ and 1.789 $\pm$ 0.002 kg $dm^{-3}$ for samples S and $S_L$, respectively.

The trend followed by the mean pore size, the amount of pores in the 10-50 nm range and the pore specific surface depends on both the product typologies and the lightweight process. In particular, the mean pore size shows the lowest values in samples S (0.19 $\pm$ 0.01 $\mu$ m) and $S_L$ (0.30 $\pm$ 0.01$\mu$ m) when compared to samples A (0.63 $\pm$ 0.01 $\mu$ m) and $A_L$ (0.42 $\pm$ 0.01 $\mu$ m). The amount of micropores follows the opposite trend, being 5.26 $\pm$ 0.05% and 3.20 $\pm$ 0.05% in the case of samples S and $S_L$ respectively, and 1.18 $\pm$ 0.01% and 2.58 $\pm$ 0.03% in the case of A and $A_L$ respectively.

The pore specific surface (PSS) is not merely related to the pore dimensions and the lightweight process seems to influence in a different way the two series of products. In fact, the PSS values of samples $A_L$ (2.8 $\pm$ 0.2 $m^2\ g^{-1}$) are higher than those measured on samples A (1.9 $\pm$ 0.2 $m^2\ g^{-1}$), while the opposite trend is shown by samples S (3.2 $\pm$ 0.3 $m^2\ g^{-1}$) and $S_L$ (2.2 $\pm$ 0.4 $m^2\ g^{-1}$).

In addition, kurtosis values, which provide a measurement of the sharpness of the pore distribution around the mean value, are in the 0-6 range for most samples, with higher values exhibited by a couple of samples in the series A and $S_L$ (table 1); hence, kurtosis does not represent a critical parameter in the comprehension of the hygrometric behaviour of clay bricks.

The binary correlations between the MEq on one side and the porosity, the mean pore size, the amount of micropores (10-50 nm) and the pore specific surface on the other side are represented in figure 2. Taking into account the MEq of ordinary (A and S) and lightweight products ($A_L$ and $S_L$) versus open porosity (figure 2A) a considerable scattering of data comes out, as expressed by the correlation coefficient ($R^2$ = 0.101). Looking more in detail at data referring to each typology, the MEq variations of samples S, and especially of $S_L$, are more pronounced and correspond to much wider porosity ranges. Moreover, in the case of samples $S_L$, notwithstanding the lightweight process does not imply a substantial change of open porosity, being the OP values in most cases superimposed to those of the ordinary products, the absorption of humidity is lower. On the contrary, the lightweight process of samples A brought about a clear increasing of the open porosity corresponding to a substantial increment of the amount of absorbed humidity.

The lacking of a clear correlation between the MEq and OP and the strong dependence on the product typologies addressed our attention towards a better understanding of the influence of shape and size of pores.

Although the results are rather dispersed ($R^2$ = 0.512) a global negative trend of MEq with the pore dimension can be found (figure 2B) so that the absorbed humidity seems much lowered when the mean pore size exceeds about 0.5 $\mu$ m. In contrast, the presence of micropores enhances the structure/water interactions, promoting the moisture absorption; this circumstance is also confirmed by the quite significant trend of MEq versus the amount of pores in the 10-50 nm range (P50): in fact, MEq goes up very quickly when the P50 values exceed about 3% (figure 2C). The lightweight products $A_L$ and $S_L$ exhibit the most scattered distribution of micropores with the MEq, contributing to the lowering of the correlation coefficient ($R^2$ = 0.612).

Among the other variables, the internal pore surface, or in other words the pore irregularities expressed by their specific surface, exhibits the most statistically significant correlation with MEq ($R^2$ = 0.718): the higher the pore specific surface, the greater the amount of absorbed moisture at equilibrium conditions (figure 2D). Once again, the main exception is represented by samples $S_L$, whose data suffer of a wider dispersion respect to the general linear trend; their MEq seems not to be merely linked to the body microstructure and pore morphology, leaving the suspect that the compositional features, including the nature of the pore-forming agent, may play a certain role.

*3.3. Predicting the MEq*

Owing to the results arising from the binary correlation of the experimental data, a different statistical approach was undertaken through multivariate analyses: principal components and multiple regression.

From a strictly statistical point of view, the principal components allow to classify all the investigated parameters into different groups, according to the correlation existing among them, and in this way to reduce the number of significant variables [18]. In our case, considering the MEq together with the main physical and microstructural parameters as variables, the analysis extracted two significant factors, accounting for 77% of the total variance. Plotting the factors in a binary diagram makes it possible to single out the relative position of each parameter and, based on their mutual distance, the most statistically significant relationships stand out. Factor 1 explains most of the variance of some physical (OP, BD) and microstructural variables (MPS, PSS and P50) while factor 2 accounts for the variance of total porosity (figure 3). The MEq is influenced mainly by the pore size, with the pore specific surface and the mean diameter working in the opposite way; moreover, the effect of the amount of micropores which influences positevely the MEq has to be claimed.

In order to quantify the influence of the microstructural parameters and trying to develop an equation modeling to predict the MEq, a stepwise multiple regression analysis was attempted. The equilibrium moisture content was taken as dependent variable, while open and total porosity, bulk density, mean pore size, pore specific surface, kurtosis and amount of micropores were considered as independent variables. The multiple regression analysis provided a forecast of the MEq value with a fair multiple correlation coefficient ($R = 0.89$, $R^2 = 0.80$) and a very good probability level (figure 4). Table 2 reports the raw (B) and standardized ($\beta$) correlation coefficients of the variables selected by the statistical procedure. According to $\beta$ values, the most effective variable is the amount of micropores, followed by the pore specific surface, while an undoubtedly minor contribution was accredited to the bulk density. For this purpose, it is important to point out that the regression procedure neglected the mean pore size, as the pore dimensions with a greatest statistical significance are below 50 nm. As a matter of fact, the capillary condensation of water vapour is expected to occur, according to Kelvin and BET laws, in the pores smaller than approximately 1 to 30 nm depending on the relative moisture [19].

The higher the amount of micropores and, consequently, their specific surface, the higher the amount of humidity which can be absorbed. The influence of bulk density is much lower and its p-level suggests a certain dependance on the sample population.

## 4. Conclusions

The equilibrium moisture content of fourty samples - representing two different typologies of ordinary (A and S) and lightweight ($A_L$ and $S_L$) clay bricks - was measured and the correlations among the MEq data and the physical and microstructural characteristics were investigated through binary and multivariate techniques (factor analysis and multiple linear regression analysis). The conclusions which can be drawn on the basis of the binary correlations are substantially confirmed by the factorial and multiple regression analyses. The porosity of clay bricks does not imply a clear correlation with the absorbed humidity and the behaviour of the products taken into account cannot be considered unambiguous, differing for both the product typology and the lightweight procedure.

The MEq is mainly influenced by the pore size, with the pore specific surface and the percentage of micropores (10-50 nm) playing a fundamental role in promoting the structure/water interactions. This circumstance is confirmed by all the different statistical approaches and it has to be properly evaluated when a structural design of the material is requested. At any event, the mean pore size which, according with the binary correlation and the factorial analysis, stood out as one of the main parameters to be considered in order to reduce the amount of absorbed moisture at equilibrium conditions has not been selected by the multiple regression analysis.

For this purpose, the results concerning the role of the pore dimensions, achieved by different approaches, indicate that the amount of humidity which can be absorbed by the clay brick is strongly reduced when:

- the mean pore size is coarser than about 0.5 $\mu$ m;
- the amount of micropores does not exceed 2%;
- the pore specific surface is smaller than about 2 $m^2$ $g^{-1}$.

The results obtained, which emphasize the role played by the pore size in respect to that exerted by the amount of porosity, represent a starting point for the microstructural design of clay bricks.

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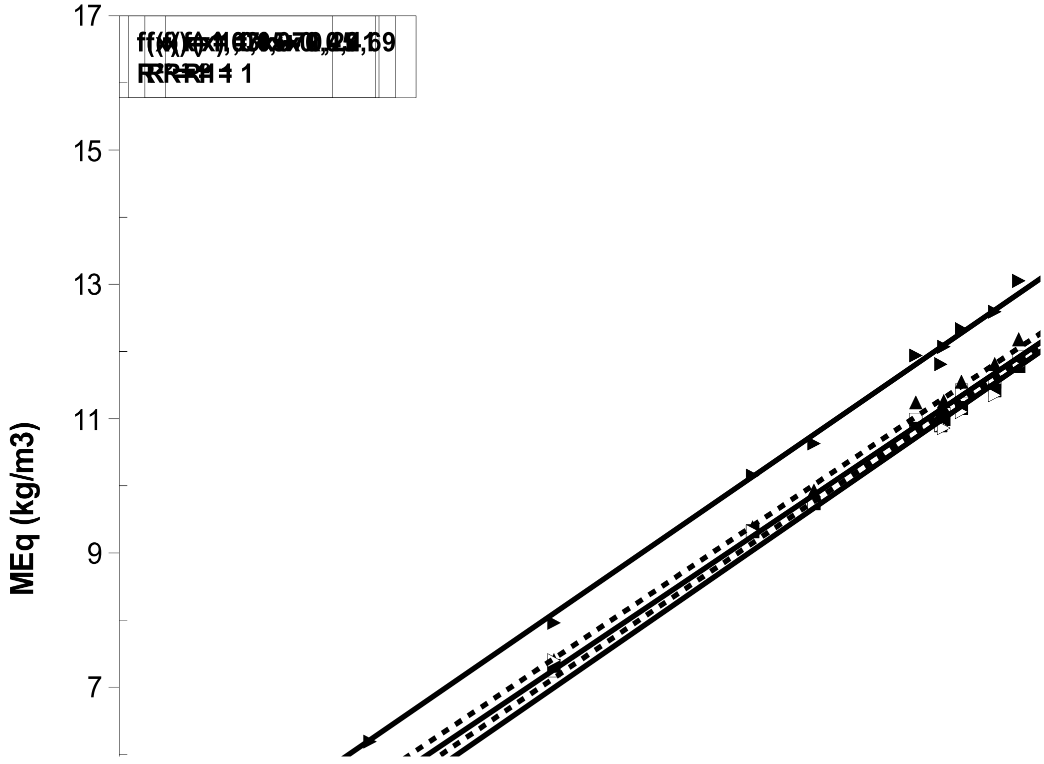


Fig. 1. Correlation of the moisture content at equilibrium (MEq) measured at different relative humidity (RH) values.

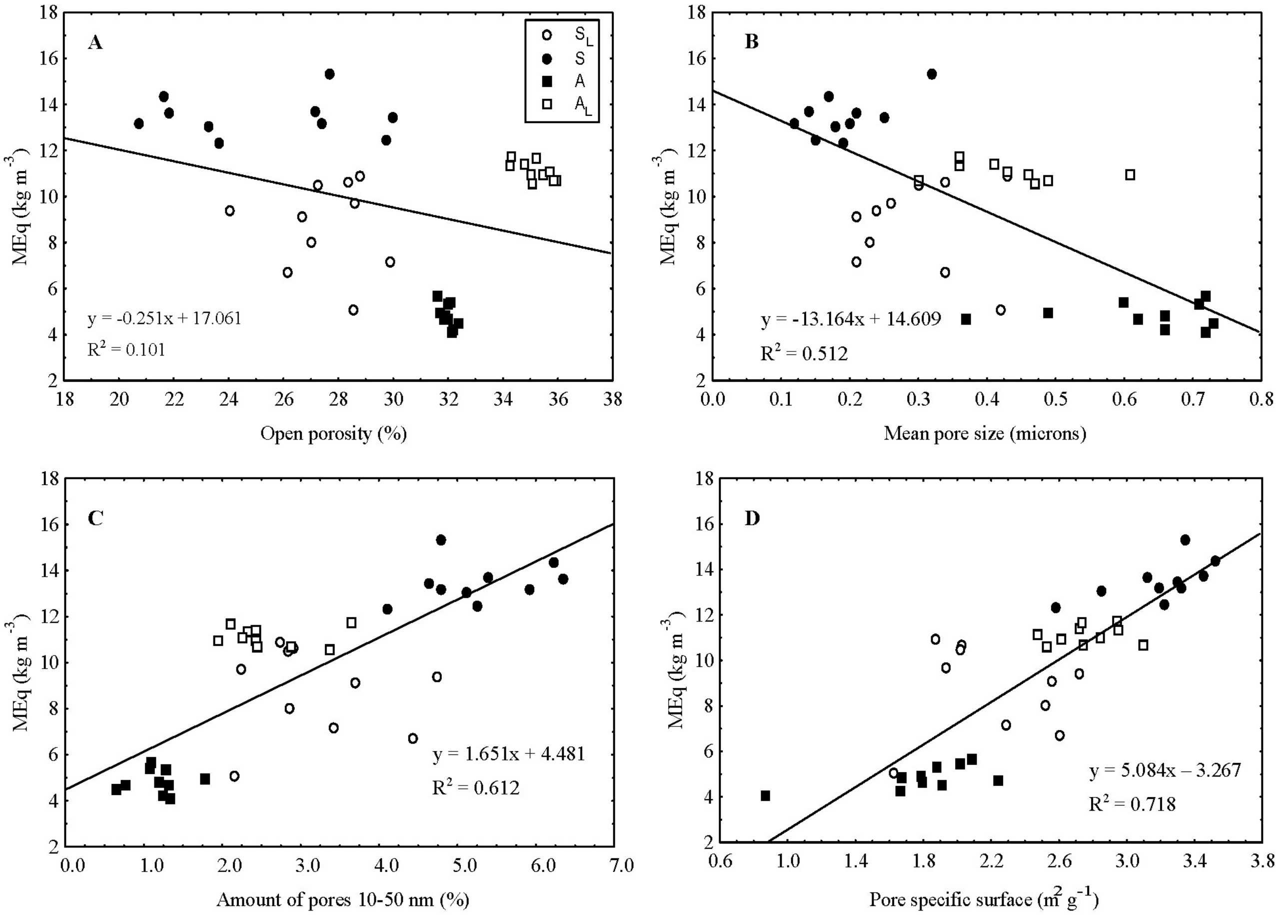


Fig. 2. Correlation of MEq with A) open porosity, B) mean pore size, C) amount of micropores, D) pore specific surface.

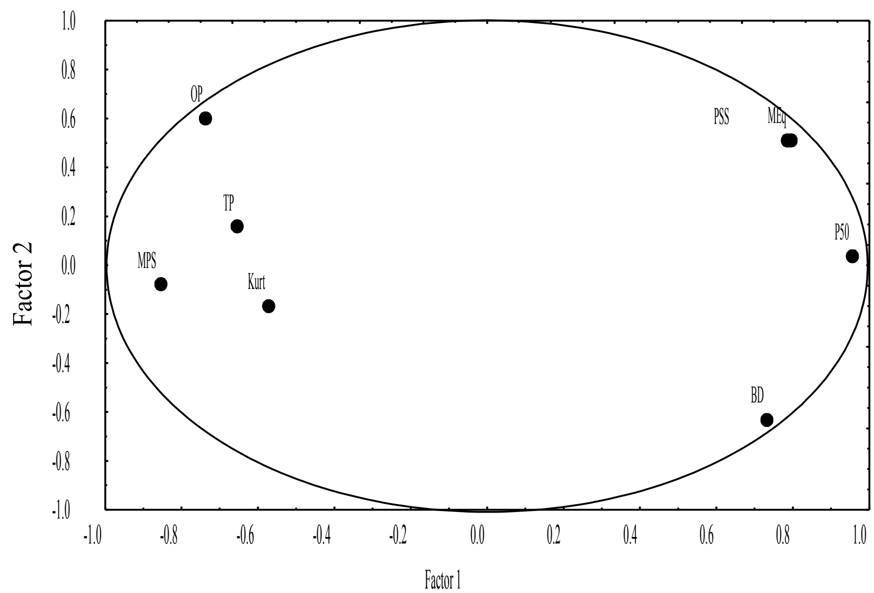


Fig. 3. Weight plot of factors 1 and 2 obtained by extraction of the main components (MEq = Equilibrium moisture content, PSS = Pore Specific Surface, P50 = Fraction of pores in the 10-50 nm range, BD = Bulk Density, MPS = Mean Pore Size, OP = Open Porosity; TP = Total Porosity, Kurt = Kurtosis of the pore size distribution).

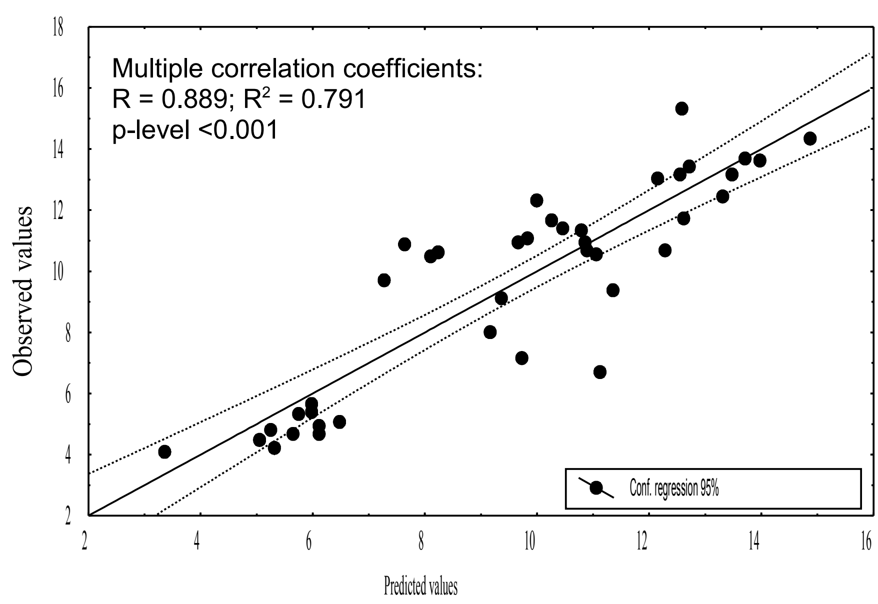


Fig. 4. Equilibrium moisture content (70% RH): observed values versus predicted values by the multiple regression analysis.

Table 1. Equilibrium moisture content (MEq), open (OP) and total (TP) porosity, bulk density (BD), mean pore size (MPS), kurtosis of pore size distribution (Kurt), percentage of pores in the 10- 50 nm range (P50) and pore specific surface (PSS) of clay bricks.

| Sample | MEq 70% RH (kg m$^{-3}$) | OP (% vol) | TP (% vol) | BD (kg dm$^{-3}$) | MPS (μ m) | Kurt | P50 (% vol) | PSS (m$^2$ g$^{-1}$) |
|---|---|---|---|---|---|---|---|---|
| $S_L$ | 10.63 | 28.4 | 35.1 | 1.777 | 0.34 | 7.29 | 2.90 | 2.03 |
| $S_L$ | 10.47 | 27.3 | 35.6 | 1.779 | 0.30 | 4.84 | 2.83 | 2.02 |
| $S_L$ | 9.38 | 24.1 | 31.9 | 1.827 | 0.24 | -0.16 | 4.74 | 2.72 |
| $S_L$ | 9.09 | 26.7 | 26.7 | 1.839 | 0.21 | 0.40 | 3.70 | 2.56 |
| $S_L$ | 10.90 | 28.8 | 30.9 | 1.777 | 0.43 | 3.75 | 2.75 | 1.87 |
| $S_L$ | 6.71 | 26.2 | 36.6 | 1.805 | 0.34 | 3.09 | 4.43 | 2.60 |
| $S_L$ | 7.18 | 29.9 | 33.7 | 1.769 | 0.21 | 1.40 | 3.42 | 2.29 |
| $S_L$ | 9.68 | 28.6 | 31.2 | 1.770 | 0.26 | 6.78 | 2.24 | 1.93 |
| $S_L$ | 8.00 | 27.0 | 33.5 | 1.789 | 0.23 | 0.56 | 2.86 | 2.52 |
| $S_L$ | 5.06 | 28.5 | 35.3 | 1.766 | 0.42 | 5.02 | 2.15 | 1.62 |
| S | 12.46 | 29.8 | 31.7 | 1.824 | 0.15 | 1.33 | 5.25 | 3.22 |
| S | 12.33 | 23.6 | 32.2 | 1.838 | 0.19 | 0.21 | 4.11 | 2.58 |
| S | 13.07 | 23.3 | 29.7 | 1.830 | 0.18 | 1.48 | 5.12 | 2.85 |
| S | 15.32 | 27.7 | 33.8 | 1.846 | 0.32 | -0.47 | 4.79 | 3.34 |
| S | 14.36 | 21.7 | 30.2 | 1.849 | 0.17 | 0.09 | 6.24 | 3.52 |
| S | 13.65 | 21.9 | 25.7 | 1.853 | 0.21 | -0.28 | 6.35 | 3.12 |
| S | 13.69 | 27.2 | 27.2 | 1.842 | 0.14 | 1.14 | 5.39 | 3.45 |
| S | 13.20 | 20.7 | 30.1 | 1.858 | 0.20 | -0.55 | 5.93 | 3.19 |
| S | 13.20 | 27.4 | 27.6 | 1.845 | 0.12 | 2.64 | 4.79 | 3.32 |
| S | 13.43 | 30.0 | 31.9 | 1.824 | 0.25 | -0.18 | 4.64 | 3.30 |
| A | 4.08 | 32.2 | 34.3 | 1.775 | 0.72 | 4.44 | 1.33 | 1.87 |
| A | 5.64 | 31.6 | 35.4 | 1.778 | 0.72 | 3.26 | 1.10 | 2.09 |
| A | 4.91 | 31.7 | 34.5 | 1.780 | 0.49 | 4.66 | 1.78 | 1.79 |
| A | 4.84 | 31.9 | 39.4 | 1.771 | 0.66 | 1.96 | 1.21 | 1.67 |
| A | 5.41 | 32.1 | 33.4 | 1.768 | 0.60 | 4.79 | 1.08 | 2.02 |
| A | 4.66 | 32.0 | 36.3 | 1.773 | 0.37 | 7.24 | 1.32 | 1.79 |
| A | 5.32 | 32.0 | 33.6 | 1.777 | 0.71 | 3.73 | 1.29 | 1.88 |
| A | 4.22 | 32.2 | 32.2 | 1.769 | 0.66 | 6.77 | 1.25 | 1.67 |
| A | 4.71 | 31.9 | 34.9 | 1.769 | 0.62 | 10.36 | 0.77 | 2.25 |
| A | 4.50 | 32.4 | 33.4 | 1.772 | 0.73 | 1.75 | 0.64 | 1.91 |
| $A_L$ | 11.33 | 34.3 | 36.6 | 1.726 | 0.36 | 0.37 | 2.33 | 2.95 |
| $A_L$ | 10.93 | 35.5 | 35.5 | 1.715 | 0.46 | 0.97 | 1.95 | 2.61 |
| $A_L$ | 11.42 | 34.8 | 35.1 | 1.720 | 0.41 | 3.09 | 2.42 | 2.72 |
| $A_L$ | 11.65 | 35.2 | 37.8 | 1.710 | 0.36 | 3.05 | 2.11 | 2.73 |
| $A_L$ | 10.97 | 35.0 | 36.1 | 1.716 | 0.61 | -0.55 | 2.44 | 2.85 |
| $A_L$ | 10.66 | 35.9 | 36.2 | 1.702 | 0.49 | 1.26 | 2.44 | 2.74 |
| $A_L$ | 10.57 | 35.1 | 35.1 | 1.729 | 0.47 | 0.57 | 3.37 | 2.53 |
| $A_L$ | 10.68 | 35.8 | 37.6 | 1.704 | 0.30 | 13.89 | 2.88 | 3.10 |
| $A_L$ | 11.70 | 34.3 | 36.4 | 1.720 | 0.36 | 1.64 | 3.65 | 2.94 |
| $A_L$ | 11.10 | 35.7 | 35.7 | 1.622 | 0.43 | 0.93 | 2.26 | 2.47 |

Table 2

Results of the multiple regression analysis. The multiple correlation coefficient (R and $R^2$), the standardized (β ) and the raw regression coefficient (B), with their relative errors, as well as the probability level (p) for each variable selected are presented.

| Samples = 40 | Standardized regression coefficients | | Raw regression coefficients | | Probability level |
|---|---|---|---|---|---|
| | β | standard error | B | standard error | p |
| Intercept | | | 35.140 | 14.997 | 0.024 |
| PSS | 0.445 | 0.136 | 2.456 | 0.754 | 0.002 |
| P<50 | 0.627 | 0.176 | 1.323 | 0.371 | 0.001 |
| BD | -0.280 | 0.117 | -20.100 | 8.346 | 0.021 |
| Multiple correlation coefficients: R = 0.889 ; $R^2$ = 0.791 | | | | | |