Thermal Conductivity of Clay Bricks

Michele Dondi¹, Francesca Mazzanti¹, Paolo Principi², Mariarosa Raimondo¹, Giorgio Zanarini³

¹CNR- Institute of Science and Technology for Ceramics, Faenza (Italy); ² Department of Energetics, University of Ancona, Ancona (Italy) ³Consorzio Alveolater, Bologna (Italy)

Abstract: In the present work the thermal conductivity of twenty-nine samples of clay bricks was measured and the correlations of the thermal performance with the compositional, physical and microstructural features of products were investigated. The results obtained directed our attention toward a better understanding of the role played by some parameters (i.e. mineralogical components and pore size distribution), other than bulk density, in improving or depressing the insulating properties of bricks. Among them, the unfavourable role of quartz, Ca-rich silicates and amorphous phase came out, while the role of pore size and specific surface should be more accurately evaluated in the structural design of materials.

Key Words: bricks, thermal insulation, bulk density, mineralogy, microstructure.

1. Introduction

Due to the ever increasing requirements for energy saving and a pressing competition with alternative building materials, the thermal insulating properties of clay bricks have recently become more and more important (Krahl 1989). Many studies have been devoted to better understand the way of improving the thermal performance of clay bricks, acting on both the physical properties of terracotta (porosity, etc.) and the geometrical design of products (Hauck et al. 1998, Rimpel and Schmedders 1996, Jungk et al. 1997, Schmidt-Reinholtz 1990, Jungk and Krcmar 1996, Anton 1993, Krahl 1989).

These studies point out that the thermal conductivity of bricks is mainly related to their bulk density, so that increasing the thermal insulating properties implies the production of materials with a higher porosity (Jungk et al. 1997, Schmidt-Reinholtz 1990, Jungk and Krcmar 1996). However, the correlation between thermal conductivity and bulk density is not statistically significant since data exhibit on the whole a considerable scattering (Fig. 1). As a matter of fact, bulk density alone is not able to describe and accurately reflect the thermal behaviour of clay bricks. The different analytical methods used to measure the thermal conductivity (Anton 1993, DIN 4108, UNI 1994, Albenque 1992) probably account for some discrepancies but, in most cases, the compositional and microstructural features of bricks play a very important role (Jungk and Krcmar 1996, Dondi et al. 2000, Schulle and Kutzendorfer 1988, Schlegek et al. 1999, Rimpel and El Ghazzali 1998).

This work is aimed at outlining the thermal conductivity of clay bricks trying to single out the compositional, physical or microstructural parameters which affect their thermal behaviour most significantly. Moreover, a statistical treatment of data was performed in order to quantify the influence of the above mentioned characteristics on thermal conductivity.

2. Materials and Methods

Twenty-nine samples of clays, collected in twenty-one different brickworks, were selected in order to represent the wide range of raw materials utilized by the Italian brick industry (Fabbri and Dondi 1995). The sampling procedure and the manufacturing of clay brick elements suitable for the conductivity measurements were described in details in previous work (Dondi et al. 2000). Sampling was carried out in every brickwork, after clay treatment and extrusion, while drying and firing were performed in a single industrial plant. After firing, all products were ground and polished to get the suitable geometry (disks of $200 \pm 1 \text{ mm}$ diameter, $20 \pm 2 \text{ mm}$ of thickness, 0.05% planarity) for thermal conductivity measurements.

The thermal conductivity of bricks was measured by the hot plate method, according to UNI 7745 (1977) Standard with a Dynatech TCFGM apparatus. For each typology of product, 6 specimens were tested with an experimental uncertainty lower than 0.5%.

Phase composition, open, closed and total porosity, bulk density, pore size distribution and pore specific surface were determined on fired products.

The phase composition was quantitatively determined by X-ray powder diffraction (Rigaku Miniflex, CuK α radiation) with the Reference Intensity Ratio method (Al₂O₃ as internal standard). The experimental error is within 5% relative.

Open porosity (OP) and bulk density were quantified by measuring dry weight, watersaturated weight and the weight suspended in water, according to ASTM C 373 (1994). Total porosity (TP) was calculated as the ratio between bulk density and specific weight according to ASTM C 329 (1994); the amount of closed porosity (CP) was estimated by the difference: CP = TP - OP.

The pore size distribution (in the 0.01 – 100 μ m range) was determined by mercury intrusion porosimetry (Carlo Erba Porosimeter 2000) with an experimental uncertainty of about 1% relative. The pore specific surface analysis was performed by nitrogen absorption (Micromeritics FlowSorb II 2300) following the B.E.T. (Brunauer, Elmet, Teller) single point method according to ASTM C 1069 (1997).

A statistical elaboration of data was performed by simple (linear binary correlation) and multivariate analysis techniques (factor analysis, multiple linear regression analysis and structural equation modeling) using the StatSoft Statistica 5.0 software. Factor analysis was carried out on the main physical, compositional and microstructural variables extracting principal components (4 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

On the whole, the samples considered here showed a great variability of thermal (Table 1), compositional (Table 2), physical and microstructural parameters (Table 3), giving a significant survey of the Italian clay brick production.

A comparison of the relationship between the thermal conductivity data collected from the literature and those obtained in the present work with the bulk density is reported in Figure 2. The existence of a correlation for both series of data with bulk density is quite evident, but it is very clear that the presence of some relevant exceptions addresses our attention toward a better understanding of the other variables which can affect the thermal conductivity values. In fact, in some cases, for the same bulk density we have products showing very different thermal behaviour, probably linked in a more complex way to the microstructure. Owing to these results, it is reasonable to suppose that not only porosity, and hence bulk density, but also the size and shape of pores (Schulle and Kutzendorfer 1998, Schlegek et al 1999), as well as the presence of a certain mineralogical component (Rimpel and El Ghazzali 1998), can play a very important role. Moreover, this thesis is supported by more detailed studies of the heat transfer mechanism, which can be enhanced or depressed, for example, by the different free mean path of air molecules entrapped into the pores and, consequently, by the different pore size (Schlegel et al. 1999).

In order to shed light on the complex relationship between thermal conductivity and bulk density, as well as on the role of microstructural features and mineralogical composition, we performed a statistical study of the results through different approaches (binary and multiple regression analysis, factorial analysis).

The mutual correlations between thermal conductivity and the microstructural (Fig. 3) or mineralogical parameters (Fig. 4) were first evaluated. The results obtained provide just some trends, such as the vague positive correlation of thermal conductivity with bulk density or the negative correlations with open porosity and mean pore size. In the other cases, no significant relationship is detectable. Plotting the thermal conductivity against the content of single mineralogical components reveals a different role played by K-feldspar and wollastonite in improving the thermal insulating properties of bricks, in contrast to the opposite tendency exhibited by plagioclase, pyroxene and illite-mica (Fig. 4).

As far as the influence of the microstructural variables (bulk density, open, closed and total porosity, mean pore size, pore selection and specific surface), the total porosity, rather than bulk density, appears to be the only one with a statistically significant relation with thermal conductivity (Fig. 3). Some samples with a higher total porosity actually have poorer thermal insulating properties, probably because of the contrasting influence exerted by pores having different size.

The factorial analysis, performed through the analysis of the main components, confirms that the thermal conductivity is influenced by several variables and that it is quite difficult to point out the more significant parameters, together with porosity, on bulk density.

From a strictly statistical point of view, the analysis of the main components allows us to classify all the parameters into different groups, according to the correlation existing among them, and to reduce the number of significant variables (Cooley and Lohnes 1971).

In our case, the principal components analysis extracted four significant factors accounting for 67% of the total variance. Plotting the factors it is possible to single out the relative position of each parameter and, based on their mutual distance, the most significant chemical and physical analogies stand out (Fig. 5 - 6). As shown in the square of figure 5, where the generic variables A, B, C and D are represented, A is positively correlated with D, negatively correlated with C, while there is no correlation between A and B (Cooley and Lohnes 1971).

Factor 1 explains most variance of some compositional (calcium silicates, quartz), physical (closed porosity) and microstructural variables (pore size selection); factor 2 explains the variance of bulk density versus open and total porosity (Fig. 5). Factors 3 and 4 account for the variance of some phases (mica, pyroxene, hematite) as well as some microstructural parameters (mean pore size and pore specific surface) (Fig. 6).

The thermal conductivity is influenced in a complex way by many variables and the four factors are able to explain no more than half of its variance. However, a certain effect of bulk density, open and total porosity can be claimed, as the role of pore size and phase composition also stands out.

A further statistical analysis was performed by a stepwise multiple regression, taking the thermal conductivity as dependent variable and the main compositional and physical parameters as independent ones; this procedure selects progressively the independent parameters with the greater significance. Once two outliers were eliminated (sample X and WPS), the multiple regression provided a forecast of the thermal conductivity value with a fair multiple correlation coefficient (R = 0.780, R² = 0.608) with a probability level that is quite good (p<0.003) (Fig. 7). Table 4 reports the raw (B) and standardized (β) correlation coefficients of the variables selected, which are the open porosity and the amount of quartz, and Ca-rich silicates (wollastonite and melilite). Among these parameters, the more effective, according to β values, is open porosity, followed by quartz, wollastonite and melilite, though the p-level of calcium silicates is rather high, suggesting a certain dependance on the sample population. While the influence of open porosity on thermal conductivity is inversely proportional, the effect of quartz, wollastonite and melilite is opposite: the higher their amount, the higher the thermal conductivity.

In light of the results obtained, we can point out that the multiple regression confirms the indications obtained by the previous statistical approaches, although it is quite difficult to elaborate a prediction model on the basis of the β values.

4. Conclusions

In order to better understand the ways to improve the thermal performance of clay bricks, the thermal conductivity of twenty-nine samples of clay bricks was determined and the relationships with their compositional, physical and microstructural features were evaluated.

A comparison of the correlation between the thermal conductivity data collected from the literature and those obtained in the present work with the bulk density highlighted that the dependance of thermal conductivity on bulk density, quoted by several authors, is not always very obvious and that this latter parameter alone is not able to describe and accurately comprehend the thermal behaviour of clay bricks.

Through a statistical treatment of data, some trends regarding the relationships among the thermal conductivity and the main mineralogical and microstructural variables of bricks were revealed. The simple linear binary correlations and the multivariate analyses (factor analysis and multiple linear regression analysis) highligthed the role played by some mineralogical components, in particular Ca-rich silicates (wollastonite and melilite), quartz and amorphous, in depressing the insulating properties of clay bricks. On the other hand, among the microstructural parameters, the role of open porosity in improving the thermal performances of bricks is predominant, but, in many cases, the role of pores size and specific surface should be more accurately evaluated in the structural design of materials.

Appendix I. References

Albenque, M. (1992). "Thermal conductivity of ceramic bodies of fired clay – Recent measurement results". *Ziegelindustrie Int.*, 2, 93-96.

Anton, H (1993). "Thermal conductivity of brick masonry. Simplification of the measurement method for the quality control". *Ziegelindustrie Int.*, 6, 262-268.

ASTM C 329 (1994). "Specific gravity of fired ceramic whiteware materials". American Society for Testing and Materials.

ASTM C 373 (1994). "Water absorption, bulk density, apparent porosity, and apparent specific gravity of fired whiteware products". American Society for Testing and Materials.

ASTM C 1069 (1997). "Specific surface area of alumina or quartz by nitrogen adsorption". American Society for Testing and Materials.

Cooley, W. W., Lohnes, P. R. (1971). "Multivariate data analysis". New York: Wiley

DIN 4108 Part 5. "Thermal insulation in building; calculation methods". Deutsches Institut für Normung.

Dondi, M., Principi, P., Raimondo, M., Zanarini, G. (2000). "The thermal conductivity of bricks produced with Italian clays". *L'Industria dei Laterizi*, 65, 309-320.

Fabbri, B., Dondi, M. (1995). "La produzione del laterizio in Italia". Gruppo Editoriale Faenza Editrice, 1-160.

Hauck, D., Ruppik, M., Hörnschemeyer, S., Richter, F. (1998). "Influence of raw materials composition on the strength and thermal conductivity of vertically perforated clay bricks and blocks". *Ziegelindustrie Annual*, 54-80.

Jungk, K., Halseband, H., Ebert, K., Krcmar, W. (1997). "Porosifiers for the production of lightweight bricks". *Tile & Brick Int.*, 13 (1), 7-12.

Jungk, E. K. and Krcmar, W. (1996). "Reduction of the thermal conductivity of clay masonry units by optimization of the ceramic body structure". *Ziegelindustrie Int.*, 6, 368-376.

Krahl, G. (1989). "Use of masonry bricks with more stringent requirements for thermal and acoustic insulation and fire protection". *Ziegelindustrie Int.*, 12, 702-707.

Rimpel, E., Schmedders, T. (1996). "Industrial production of high-porosity brick materials". *Ziegelindustrie Int.*, 174-207.

Rimpel, E., El Ghazzali, S. M. (1998). "Influence of mineral constituents and equilibrium moisture on the thermal conductivity of bricks". *Ziegelindustrie Annual*, 28-52.

Schlegek, E., Haussler, K., Seifert, H., Freiberg, K. (1999). "Microporosity and its use in highly efficient thermal insulating materials". *cfi/Ber. DKG* 76(8), 7-10.

Schmidt-Reinholtz, C. (1990). "Suggestion for the reduction of body bulk density through additives". *Tile & Brick Int.*, 6 (3), 23-27.

Schulle, W., Kutzendorfer, J. (1988). "Relationship of the thermal conductivity and strength to the mean pore diameter in chamotte lightweight refractory blocks". *Ziegelindustrie Int.*, 6, 300-303.

UNI 10355 (1994) "Thermal resistance values and calculation method". Ente Nazionale Italiano di Unificazione.

UNI 7745 (1977). "Determinazione della conduttività termica con il metodo della piastra calda con anello di guardia". Ente Nazionale Italiano di Unificazione.

Sample	Thermal conductivity
	$W \cdot m^{-1} \cdot K^{-1}$
Х	0.63
MO	0.49
MA	0.49
AT	0.50
SL	0.47
А	0.53
SA	0.48
CA	0.52
S	0.56
F	0.46
LM	0.47
D	0.49
LS	0.49
WSP	0.60
WSN	0.39
WPP	0.54
WPN	0.42
CEP	0.54
CEN	0.50
CAP	0.44
CAN	0.46
	0.44
	0.42
	0.46
	0.48
RUB	0.46
	0.41
	0.52
ILP	0.03

Table 1 – Thermal conductivity of clay bricks

Sam ple	Quar tz	Plagiocl ase	K_felds par	Pyroxe ne	Wollasto nite	Melil ite	Hema tite	Illite/ mica	Calci te	Pericla se	Amorph ous
Х	20	20	7	6	4	16	0	11	0	0	16
MO	26	11	4	8	10	8	2	0	0	0	31
MA	42	11	11	2	0	0	0	6	0	0	28
AT	28	21	8	0	3	12	1	20	0	0	7
SL	26	2	8	0	0	0	1	20	0	0	43
А	22	23	11	6	6	9	1	12	0	0	10
SA	22	23	9	14	0	11	1	9	2	0	9
CA	38	9	17	2	3	5	3	12	0	0	11
S	26	11	14	0	0	0	4	0	0	0	45
F	18	25	13	7	2	2	4	0	0	0	29
LM	26	18	3	2	1	8	0	23	0	0	19
D	16	12	5	4	7	30	1	0	1	0	24
LS	20	10	5	15	8	27	2	8	1	0	4
WSP	38	8	7	8	0	6	1	5	0	1	26
WSN	31	21	3	9	5	6	2	4	0	3	16
WPP	40	15	2	12	0	9	2	4	1	2	13
WPN	38	18	8	17	0	6	1	0	0	0	12
CEP	22	20	9	8	9	14	0	5	0	0	13
CEN	24	23	4	11	8	14	1	7	0	0	8
CAP	36	11	6	4	4	12	3	13	2	3	6
CAN	25	13	4	6	7	6	4	11	0	4	20
ATP	27	23	3	3	4	16	2	13	0	3	6
ATN	20	19	8	6	4	14	3	10	0	5	11
RIP	33	13	9	2	2	0	3	2	0	0	36
RIN	45	13	5	8	0	2	3	2	0	0	22
RDB	21	22	2	8	11	5	1	16	0	0	14
RIX	39	7	4	2	2	2	4	11	0	0	29
ILS	17	13	2	9	8	14	2	11	0	2	22
ILP	35	16	3	7	7	4	5	0	0	0	23

Table 2- Phase composition of clay bricks (wt %)

Table 3 – Open (OP), closed (CP) and total (TP) porosity, bulk density (BD), mean pore size (MPS), pore size selection (PSE), pore specific surface (PSS) and fraction of pores smaller than 50 nm (P<50) of clay bricks

Sampl	OP	СР	TP	BD	MPS	PSE	PSS	P<50
е	(% vol)	(% vol)	(%vol)	(kg m ⁻ °)	(µm)	(adim)	(m² g⁻¹)	(%)
Х	34.8	1.9	36.7	1.76	0.7	0.3	1.6	0.8
MO	36.3	0.2	36.5	1.77	0.7	0.4	1.2	0.9
MA	30.5	0.0	30.5	1.87	0.4	0.7	8.3	11.3
AT	39.0	3.6	42.6	1.61	0.8	0.4	2.4	1.8
SL	28.9	0.0	28.9	1.90	0.1	0.6	12.3	1.2
А	25.2	6.6	31.8	1.85	0.5	0.3	1.4	1.5
SA	27.2	7.3	34.5	1.81	0.7	0.3	1.3	1.0
CA	33.3	1.6	34.9	1.76	1.1	0.4	2.0	1.6
S	18.8	0.0	18.8	2.12	1.0	0.4	0.6	17.5
F	24.4	2.8	27.3	1.87	0.7	0.3	1.0	1.2
LM	27.9	5.0	32.9	1.79	0.6	0.4	1.6	1.9
D	32.7	7.8	40.5	1.67	0.4	0.2	2.2	1.5
LS	33.6	5.9	39.4	1.70	0.8	0.3	1.4	1.2
WSP	33.3	1.5	34.8	1.73	0.6	0.8	3.2	6.3
WSN	35.2	2.9	38.1	1.67	1.4	0.5	1.4	1.3
WPP	36.5	2.1	38.6	1.66	1.6	0.6	1.6	2.1
WPN	36.6	2.6	39.2	1.65	1.2	0.6	1.5	1.6
CEP	27.9	9.6	37.4	1.71	0.5	0.6	1.7	2.9
CEN	28.0	7.5	35.5	1.72	0.7	0.5	1.5	0.9
CAP	36.7	1.7	38.4	1.68	1.0	0.3	1.9	0.9
CAN	36.0	3.3	39.3	1.70	1.0	0.4	2.0	1.2
ATP	38.4	3.0	41.4	1.63	0.9	0.4	2.4	1.6
ATN	38.5	2.5	41.0	1.63	0.9	0.4	2.3	1.2
RIP	32.7	1.3	34.0	1.72	1.6	0.7	1.5	4.8
RIN	33.3	2.8	36.2	1.71	1.6	0.7	2.1	4.1
RDB	36.1	2.9	39.0	1.65	0.8	0.4	1.7	1.1
RIX	37.4	0.0	37.4	1.66	0.8	0.7	5.6	7.8
ILS	27.7	9.8	37.5	1.72	0.6	0.3	1.9	2.0
ILP	23.4	5.8	29.1	1.87	1.0	0.7	1.0	3.9

Table 4 – Results of the multiple regression analysis. The multiple correlation coefficient (R and R²), 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.

N = 27	Standardize coeff	ed regression icients	Raw re coeff	gression icients	Probability level				
	β	standard	В	standard	р				
Intercept			0.603	0.047	0.000				
Open porosity	-0.882	0.157	-0.008	0.001	0.000				
Wollastonite	0.423	0.166	0.006	0.002	0.018				
Quartz	0.600	0.195	0.004	0.001	0.005				
Melilite	0.330	0.178	0.002	0.001	0.077				
Multiple correlation coefficients: $R = 0.780$; $R^2 = 0.608$									



Figure 1. Thermal conductivity vs bulk density values.



Figure 2. Thermal conductivity vs bulk density: values collected from the literature and those obtained in the present work.



Figure 3. Thermal conductivity vs a) open porosity, b) total porosity, c) bulk density, d) mean pore size, e) pore specific surface and f) pore size selection.



Figure 4. Thermal conductivity vs a) K-feldspar, b) wollastonite, c) plagioclase, d) pyroxene, e) illite-mica content.



Figure 5. Weight plot of factor 1 and 2 obtained by extraction of the main components (PSE=pore size selection; MPS=mean pore size; PSS=pore specific surface; P50=fraction of pores smaller than 50 nm; OP=open porosity; TP=total porosity; CP=closed porosity; BD=bulk density; Qz=quartz; AM=amorphous; KF=K-feldspar; HE=hematite; PE=periclase; IM=illite-mica; CC=calcium carbonate; CS=calcium silicates; PX=pyroxene; PL=plagioclase; ME=melilite; WO=wollastonite; TC=thermal conductivity).



Figure 6. Weight plot of factor 3 and 4 obtained by extraction of the main components (PSE=pore size selection; MPS=mean pore size; PSS=pore specific surface; P50=fraction of pores smaller than 50 nm; OP=open porosity; TP=total porosity; CP=closed porosity; BD=bulk density; Qz=quartz; AM=amorphous; KF=K-feldspar; HE=hematite; PE=periclase; IM=illite-mica; CC=calcium carbonate; CS=calcium silicates; PX=pyroxene; PL=plagioclase; ME=melilite; WO=wollastonite; TC=thermal conductivity).



Figure 7. Thermal conductivity: observed values vs predicted ones.