11DBMC International Conference on Durability of Building Materials and Components
ISTANBUL, Turkey 11-14 May 2008

Durability of Clay Roofing Tiles: Assessing the Reliability of Prediction Models

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ABSTRACT

The indirect prediction of frost resistance of construction materials has been widely investigated in recent years giving rise to several models based on pore size, open porosity, water absorption and/or capillary rise. This study is aimed at appraising these models on 13 industrially-manufactured roofing tiles with a different frost resistance determined by severe freeze/thaw testing (EN 539-2). Samples were characterised by measuring pore size distribution (MIP), pore specific surface (BET), open porosity and water absorption (ASTM C67 and C373), capillary rise (UNI 10859) and phase composition (Rietveld-XRPD). No model is able to foresee reliably the product frost resistance since all models exhibit a strong dependence on the data population and probably succeed only with a homogeneous sample in terms of both composition and manufacturing technology. Among them, the Arnott's model seems to be the most reliable to discriminate among scarcely (<100 freeze/thaw cycles) and highly frost resistant products (>250 cycles); however, a complete understanding of the excellent performance (>400 cycles) provided by some roofing tiles is still lacking. Looking at their phase composition, tiles with the best performance contain also abundant calcium-magnesium silicate phases formed during firing while products with a frost resistance lower than expected are characterised by large amounts of amorphous phase or residual mica-illite and quartz. This circumstance indicates new ways to achieve highly frost resistant products that is alternative to the conventional design imposing microstructural rearrangements (low porosity and coarse pore size) through drastic batch changes and/or firing at higher temperature.

KEYWORDS

Roofing tiles, Frost resistance, Microstructure, Phase composition, Clay.

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1 INTRODUCTION

Nowadays clay roofing tiles are widely used as exterior masonry components and their durability, defined as the ability to withstand adverse climatic conditions, is one of the most important requirements to be considered in the structural design of modern buildings.

The deterioration of construction materials can be due to several factors; among them, design and construction techniques, material properties and environmental conditions, which may be, in most cases, considered more predominant than others. Particularly, in cold regions the service lifetime of masonry components is heavily affected by the frost action and salt crystallization.

Frost action is produced when the external temperature falls below 0°C and the water included in the porous structure starts freezing; the density change of the liquid-solid water transition involves the development of an internal pressure, leading to the formation of micro-cracks whose extent can overcome the strength resistance of the material and promote inescapable damage. The extent of these damages will be strictly dependent on the exposed surface area, the saturation degree of the material, as well as on the number and size of pores. Specifically, for pore dimensions greater than a critical value and/or for a low saturation degree, the developed pressure, and hence damage, will be negligible since the free space in the pores accommodates the expansion of the freezing water; in contrast, severe structural damage can occur. Owing to these circumstances, the material characteristics, which in turn depend on the raw material formulations, the forming process, and the firing cycle, become key factors in the evaluation of the deterioration risk.

Many papers in the field of civil engineering analyse the factors affecting the durability of construction materials going also through the elaboration of prediction models. In the last years, for example, Maage [Maage [1990], Arnott [Arnott [1990], Franke and Bentrup [Franke and Bentrup è1993], Koroth [Koroth *et al.* [1998], Robinson [Robinson *et al.* [1995] and Vincenzini [Vincenzini [1974] elaborated different models which basically relate to technological (i.e., water absorption in different experimental conditions, capillary coefficient) or microstructural (i.e., porosity amount, size, and internal specific surface of pores) properties, deriving then equations able to define the product durability on the basis of the calculated indices.

This study is aimed at appraising these models on industrially-manufactured roofing tiles with varying frost resistance, experimentally determined by severe freeze/thaw testing, assessing both their reliability and restrictions. The role played by phase composition on products durability has been also analysed.

2 MATERIALS AND METHODS

Thirteen (13) industrially-manufactured roofing tiles, obtained from different working plants, were selected. On the basis of different shape and colour, they have been classified as *Marseilles* (samples A, C, F, H and L), *Portuguese* (samples B, D, E, G, I, J and M) and *Coppo* (sample K) tiles. All were produced by extrusion and fired at a maximum temperature between 850 and 1150°C with a thermal cycle of 24-48 hours from cold -to -cold.

Products were characterized by the determination of phase composition, open, closed and total porosity, bulk density, pore size distribution and pore specific surface. Phase composition (Table 1) was determined by X-ray powder diffraction (Rigaku DIII, experimental conditions: monochromated $CuK_{1,2\ \alpha}$ radiation in the 5-80°2θ range, scan rate 0.02 °2θ, 2 sec per step) and quantitative analyses were performed by Rietveld method using the GSAS software; the relative error is ± 5%

Open porosity (OP) and bulk density (BD) were quantified according to ASTM C373; specific weight (SW) was measured by He pycnometry (Micromeritics MVP 1305) according to ASTM C329 and total porosity (TP) was calculated by the equation: $TP = (1-BD/SW) \cdot 100$. In addition, the following porosity-related properties were also measured: 24-hours (WA_{24h}) and 4-hours (WA_{4h}) cold water absorption, 5-hours boiling water absorption (WA_{5h}) according to UNI 8942/3; the ratio WA_{24h}/WA_{5h} being defined as the saturation coefficient C_s (Table 2).

Table 1. Phase composition of roofing tiles.

Sample	*Quartz* (%)	*K-Feldspar*	*Plagioclase* (%)	*Pyroxene* (%)	*Hematite* (%)	*Illite* (%)	*Ghelenite* (%)	*Amorphous* (%)
A	38	8	22	14	4	-	-	14
B	24	3	11	1	5	13	-	42
C	26	7	33	12	2	5	6	8
D	20	5	22	32	5	-	1	17
E	33	7	30	3	2	-	1	23
F	24	7	26	3	3	-	-	37
G	42	13	5	6	7	-	-	27
H	23	6	41	2	2	-	2	24
I	31	6	19	1	6	9	-	27
J	20	6	36	1	1	8	-	27
K	6	1	24	9	2	-	-	58
L	24	4	44	2	2	-	1	23
M	39	7	-	-	4	10	-	40

Table 2. Open (OP), total (TP) and closed (CP) porosity, bulk density (BD), water absorption (WA), 24-hours (WA_{24h}) cold water absorption, 5-hours boiling water absorption (WA_{5h}) of roofing tiles.

Sample	*OP* (%)	*TP* (%)	*CP* (%)	*BD* (g/cm^3)	*WA* (%)	WA_{24h} (%)	WA_{5h} (%)
A	20.3	30.4	10.1	1.877	10.8	10.7	15.4
B	18.0	25.1	7.1	2.020	8.9	8.8	11.1
C	27.8	32.6	4.8	1.817	15.3	14.9	16.1
D	21.0	29.6	8.6	1.899	11.1	10.6	11.4
E	13.0	29.2	16.2	1.910	6.8	8.0	13.6
F	22.3	29.1	6.8	1.912	11.7	11.1	13.0
G	19.3	25.9	6.7	1.997	9.6	10.1	12.3
H	23.2	29.6	6.3	1.899	12.2	12.2	14.3
I	21.1	27.2	6.1	1.962	10.8	11.1	13.1
J	25.2	31.0	5.8	1.861	13.5	13.1	14.7
K	24.3	29.7	5.4	1.895	12.8	11.6	14.5
L	22.4	28.6	6.2	1.924	11.7	11.6	13.5
M	19.4	23.2	3.9	2.070	9.3	9.9	11.1

The pore size distribution (in the 0.01 – 100 μm range) was determined by MIP (ThermoFinningan Pascal 140) with an experimental uncertainty of about 1% relative; the experimentally measured mercury - brick contact angle of 166.4° was inserted into the Washburn equation [34] and the porosimetric results corrected by a factor of 1.24. Data were expressed as medium pore diameter (MD), PV (cumulative volume of pores, cm^3/g), P3 (relative amount of pores having a diameter larger than 3 μm), Φ_{50} (median pore size, μm) and Φ_{90} (ninetieth percentile, μm); the pore specific surface analysis was performed by nitrogen absorption (Micromeritics FlowSorb II 2300) following the B.E.T. single point method (ASTM C1069) (Table 3).

Table 3. Medium pore diameter (MD), cumulative volume of pores (PV), relative amount of pores having a diameter larger than 3 µm (P3), Φ_{50} (median pore size), Φ_{90}, pore specific surface (BET) and capillary coefficient (K_s) of roofing tiles.

Sample	*MD* (µm)	*PV* (cm^3/g)	*P3* (%)	Φ_{50} (µm)	Φ_{90} (µm)	*BET* (m^2/g)	*Ks* ($g/cm^2\ s^{1/2}$)
A	1.88	0.146	56.0	1.8	4.5	0.728	5.8
B	0.52	0.134	3.3	0.5	1.2	1.712	2.2
C	0.41	0.189	0.7	0.4	0.8	1.808	5.2
D	0.34	0.161	1.6	0.4	0.7	1.613	2.6
E	0.70	0.151	14.2	0.5	3.0	1.110	0.1
F	0.36	0.157	2.42	0.4	0.8	1.530	4.5
G	0.74	0.132	12.9	0.7	3.5	1.565	0.8
H	0.50	0.168	1.9	0.5	1.3	1.179	4.0
I	0.40	0.146	2.4	0.4	1.2	1.841	4.8
J	0.38	0.182	3.1	0.4	0.8	1.630	4.4
K	0.41	0.179	1.4	0.4	0.7	1.312	3.0
L	0.37	0.161	2.4	0.4	0.9	1.837	3.6
M	0.34	0.117	8.6	0.3	1.4	2.190	3.3

The capillary absorption was determined according to UNI 10859 on small pieces of about 5 cm^3 obtained from each roofing tile. Plotting the specimen mass recorded after 10, 20, 30 and 60 minutes versus the square root of the elapsed time, a graph was obtained presenting an initial straight line, whose slope is the experimental capillary coefficient K_s (Table 3). Frost resistance was assessed by severe freeze/thaw testing (EN 539-2) on 10 samples for each industrially-manufactured product. Tests were carried out in a climatic cell at temperatures ranging from -15°C to +15°C performing 400 different freeze/thaw cycles; this number of cycles - much higher than those (150) scheduled by the reference standard - was chosen in order to simulate very extreme conditions.

Table 4. Frost resistance of roofing tiles determined according to EN 539-2.

Sample	*Number of freeze/thaw cycles*	*Overcame freeze/thaw cycles*	*Defects found after freeze/thaw testing*
A	400	400	Several chips on the back of the sample
B	400	375	Loss of interlocking ribs, hairline cracks
C	50	25	Large exfoliations on both sample sides
D	400	400	none
E	300	275	Several exfoliations on both sample faces
F	125	100	Loss of interlocking ribs, delaminations, flaking, hairline
G	125	100	Breaking of test sample into three pieces
H	400	400	Detachment of a sample projection
I	100	75	Exfoliations, hairline cracks
J	125	100	Delaminations, exfoliations, hairline cracks, loss of interlocking ribs
K	125	100	Surface cracks, delaminations, exfoliations, structural cracks, pits
L	300	275	Exfoliations, loss of interlocking ribs, hairline cracks
M	150	50	Loss of interlocking ribs, chips, hairline cracks, delaminations

The product durability was evaluated from the analysis of both the mass loss and the structural changes standing out during and after the test; the freeze/thaw resistance was expressed as the fraction of samples which are able to withstand test conditions (Table 4).

Both microstructural and porosity-related parameters were utilized to calculate durability indices according to the models proposed by Maage, Robinson, Vincenzini, Arnott, Franke and Bentrup and Koroth; the variables considered by the authors to elaborate their models and the resulting indices are summarized in Table 5 and 6, respectively.

Table 5. Prediction models and classification of frost behaviour as proposed by the authors**.**

Durability Factor (DF)	*Models*	*Frost resistant products*	*Non-frost resistant products*
Maage	3.2/PV + 2.4P3	DF > 70	DF < 70
Robinson	$[K_s/10(1-C_s)] + (WA_{24h} - 10)$	Low values of DF	-
Arnott	$9.2\ P3 - 0.5\ K_s + 423\ (WA_{5h}/WA) - 100\ K_s\ C_s - 84.5$	High values of DF	-
Vincenzini	Φ_{90}	$\Phi_{90} \geq 1.80$ μm	$\Phi_{90} \leq 1.80$ μm
Franke & Bentrup	Φ_{50}	$\Phi_{50} \geq 1.65$ μm	$\Phi_{50} \leq 0.60$ μm
Koroth	$450(2.94 + WA_{5h}) + 330\ (1-WA_{4h}/WA_{5h})$	DF > 85	DF < 70

Table 6. Calculated durability factors of roofing tiles.

Sample	*Durability factors*					
	Maage	*Robinson*	*Arnott*	*Vincenzini* *Φ90 (μm)*	*Franke-Bentrup Φ50 (μm)*	*Koroth*
A	156.3	0.9	962	4.5	1.8	220
B	31.8	-1.2	394	1.2	0.5	288
C	18.7	4.9	273	0.8	0.4	238
D	23.9	0.6	272	0.7	0.4	277
E	55.2	-2.0	835	3.0	0.5	355
F	26.1	1.1	321	0.8	0.4	233
G	55.0	0.1	495	3.5	0.7	328
H	23.6	2.	343	1.3	0.5	254
I	27.7	1.2	365	1.2	0.4	227
J	25.0	1.7	314	0.8	0.4	246
K	21.2	3.1	327	0.7	0.4	279
L	25.6	1.7	339	0.9	0.4	259
M	47.9	-0.1	410	1.4	0.3	255

A statistical elaboration of data was performed by simple (linear binary correlation) and multivariate analysis techniques (factor and multiple regression analyses) using the StatSoft Statistica 6.0 software. Factor analysis was carried out on the main physical, compositional and microstructural variables extracting principal components (3 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 Reliability of Models

The durability factors of table 6 account for a quite different behaviour of samples in terms of their ability to overcome frost action. In order to verify the reliability and the validity range of the models, a comparison between the experimental results – in terms of number of freeze/thaw cycles – and the calculated indices haves been undertaken (Figure 1).

As shown in Fig. 1, the following considerations can be drawn out:

- Maage durability factor (Figure1A), although presents a positive relationship with the number of freeze/thaw cycles, is not able to discriminate among frost (DF >70) and non-frost resistant (DF<70) products, with sample A being the only one that can be considered as frost resistant; in addition, the model provides no reasons for the excellent performance (>400 cycles) of samples B, D and H. Looking in detail at the microstructure of sample A, it is clear that the Maage factor mainly accounts for its quite high percentage of pores having a diameter greater than 3 μm (P3 = 56%).
- Any statistical significant correlation was detected plotting the durability factor, calculated according to Robinson's model, and the experimental behaviour of samples (Figure 1B); however, the index seems to decrease when the behaviour of products becomes better.
- The model proposed by Arnott (Fig. 1C) seems to present a good agreement with the experimental performance of scarcely resistant (<100 cycles) and highly resistant (>250 cycles) products; the main exceptions are always represented by samples L, B, H and D having excellent durability. The insertion into the multiple regression analysis of different physical and technological parameters provides a model (Table 5) where the role of each variable is quantified by a statistical different weight: the amount of greater pores (P3) seems to be the most influent so that, accordingly, index values increases as the percentage of P3 increases.
- The limit value of Φ_{90} = 1.8 μm proposed by Vincenzini to discriminate among non-frost (Φ_{90} <1.8 μm) and frost (Φ_{90} >1.8 μm) resistant products revealed in this case not to be reliable. Specifically, examining Fig 1D, the correspondence between the experimental performance and the index can be validated only for samples A and E, while sample G, although presenting a Φ_{90} value as high as 3.5 μm, does not show an acceptable durability.
- The evaluation of the median pore size values Φ_{50}, as proposed by Franke and Bentrup (Figure 1E), presents more or less the same limitations of Maage factor. In fact, A and G are the only samples which could be considered as frost resistant, though the experimental results indicate that good (samples E and L) or excellent (samples B, D, H) performance belong to products having a reduced value of pore size (Φ_{50} <0.5-0.6 μm).
- Finally, according to Koroth's model (Figure 1F), all samples should be considered frost resistant since the durability factor calculated for each of them is much higher than the limit value of 85; however, any significant relationship between the calculated index and the experimental results occurs.

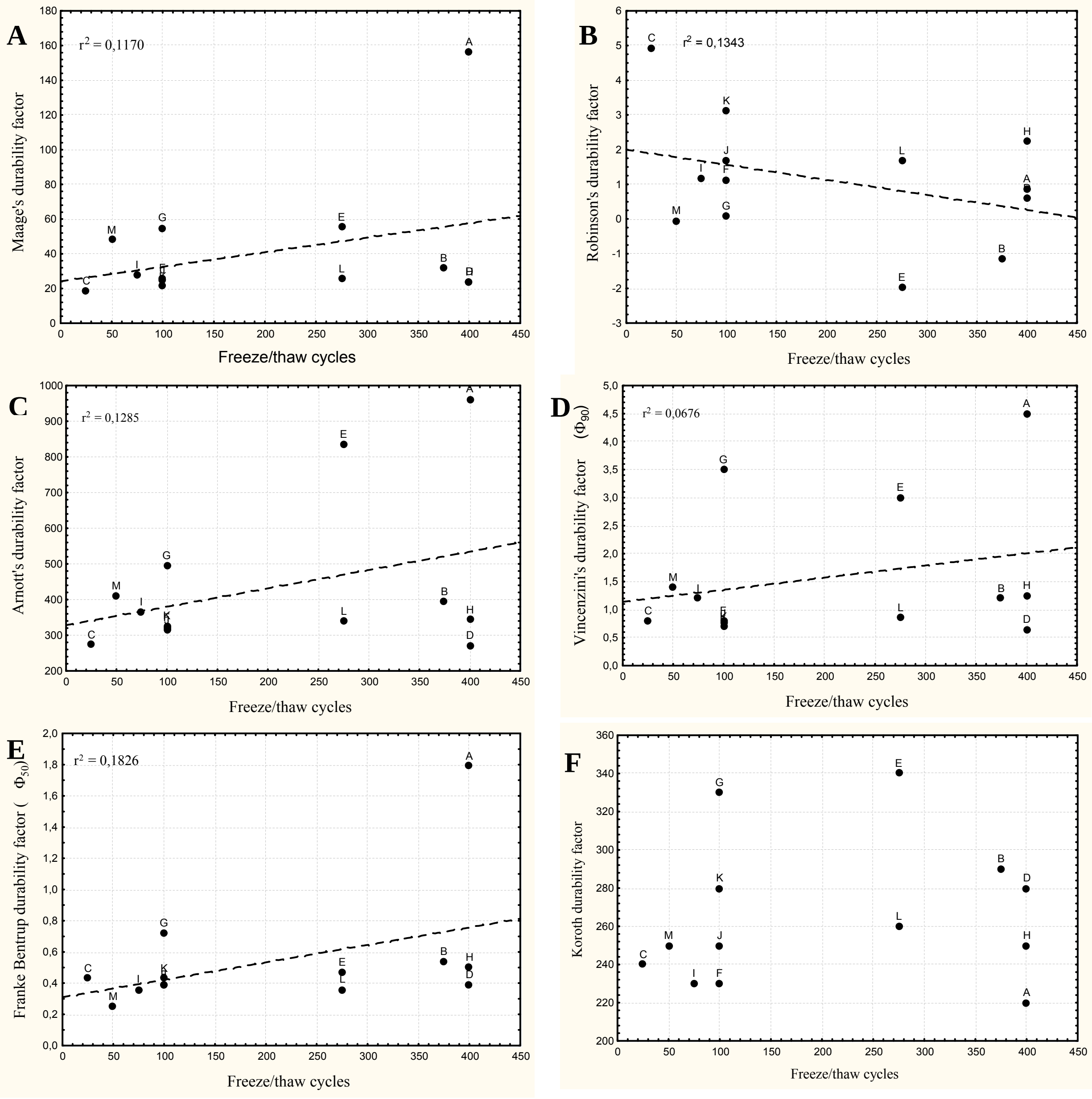

Figure 1. Experimental behaviour of roofing tiles (number of freeze/thaw cycles) vs durability factors of A) Maage; B) Robinson; C) Arnott; D) Vincenzini; E) Franke and Bentrup; F) Koroth.

3.2 Statistical Elaboration

Since simple binary correlations between the calculated indices and the experimental behaviour of roofing tiles were not satisfactory, a statistical elaboration of data (factor analysis and “stepwise” multiple regression analysis) was undertaken considering, besides the number of freeze/thaw cycles, physical (TP, BD, P3, BET, Φ_{50} and Φ_{90}) and compositional (amount of quartz, amorphous phase and Ca-silicates) properties of products as well.

- The extraction of the principal components (Table 7) highlights that, as expected, the frost resistance of products is correlated in a quite complex way with all the other variables so that the hypothesis that more than one parameter could simultaneously influence the materials real performance is in some way confirmed.

Table 7. Extraction of the principal components.

Variable	*Factorial weights*		
	Factor 1	Factor 2	Factor 3
Freeze/thaw cycles	0.535	-0.001	0.457
TP	0.545	-0.794	-0.123
P3	0.813	0.495	0.056
BET	-0.837	0.04	-0.438
BD	-0.542	0.794	0.127
Φ_{50}	0.864	0.381	0.115
Quartz	0.250	0.793	-0.528
Ca-silicates	0.457	-0.841	-0.008
Amorphous	-0.610	0.178	0.691
Φ_{90}	0.707	0.649	-0.042
Variance	4.132	3.446	1.214

Anyway, looking at the mutual relationships of figure 2, the following conclusions can be drawn out:

- a positive correlation exists between frost resistance and the amount of pores greater than 3 µm (P3), the median pore size (Φ_{50}) and Φ_{90} values;
- frost resistance is inversely related to the pore specific surface (BET) but also to the amount of amorphous phase, so that the highest its content, the lowest is the product performance;
- concerning the amount of quartz and Ca-silicates, their role on the frost resistance seems to be the opposite: a higher amount of new formed phases (such as Ca-silicates) increases the ability of products to withstand frost action, which in turn is decreased by the presence of high quartz amounts.

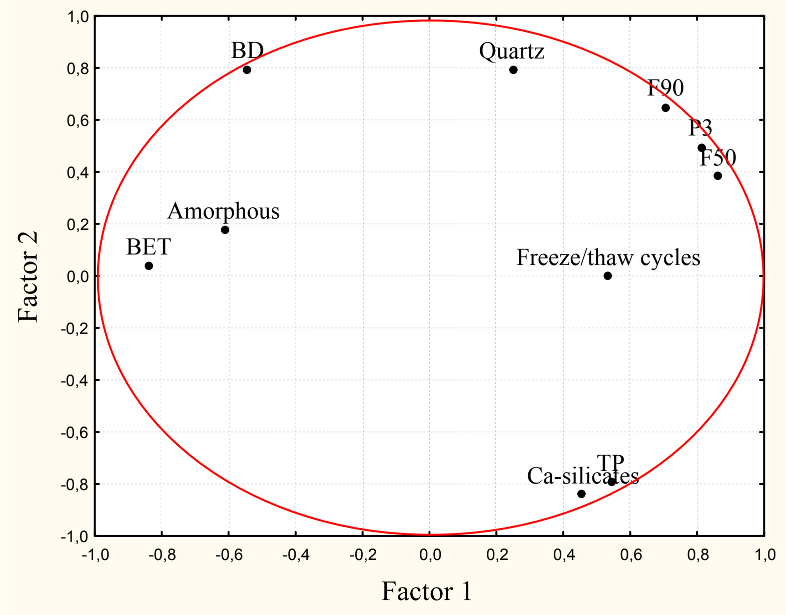

Figure 2. Extraction of the principal components: factor 1 vs factor 2.

The results of the multiple regression analysis (R^2= 0.94138, p<0.00184), taking into account the product frost resistance as dependent variable and the physical and compositional parameters as independent ones, made possible to confirm some of these indications (Table 8):

- according with the values of β factors, the statistical procedure selected the following as the most influent parameters: BD, quartz amount, BET, Ca-silicates amount, amorphous amount and Φ_{50};
- it is worth noting that the statistical weight (as expressed by β factors) of the mineralogical phases are quantitatively equivalent to that of BET and prevailing on the pore size represented by Φ_{50};
- concerning the positive or negative role played by each selected variable, it is confirmed that the amount of Ca-silicates, on one side, and that of quartz and amorphous, on the other side, work in the opposite way; a higher amount of new formed phases increases the frost resistance of products, while the presence of both quartz and amorphous potentially involves the risk of frost damage;
- the evaluation of the p-level corresponding to each selected parameters allows to assess the reliability of the results of the statistical procedure which can be considered satisfactory.

Table 8. Results of the multiple regression analysis.

Stepwise multiple regression analysis R = 0.970; R2 = 0.941; p-level < 0.002

N = 13	β	Std. Error	B	Std. Error	p-level
Intercept			-7063.22	1052.63	0.0005
BET	-0.821	0.201	-338.80	83.12	0.0065
Amorphous	-0.775	0.381	-9.20	4.53	0.0885
BD	1.857	0.231	4227.63	527.29	0.0002
Quartz	-0.882	0.368	-14.33	5.98	0.0054
Ca-silicates	0.779	0.223	7.34	3.99	0.0110
Φ_{50}	0.252	0.182	101.05	73.41	0.2180

Following these results, it can be pointed out that the production of roofing tiles with excellent frost resistance – which are able to overcome a number of freeze/thaw cycles much higher that those required by the current standard – involves the evaluation of both product and processing variables. As far as the composition of raw mixtures, their CaO content should be improved paying in the meantime great attention to the development of a highly porous microstructure and controlling the pore dimensions (to get lower BET values). Analogously, the amount of new formed Ca-silicates should be improved (≥ 40%), while the amount of amorphous phase reduced under about 20%. These requirements, together with the need to obtain a microstructure having a higher quantity of pores greater than 3 μm, can be satisfied through the modification of the firing process, i.e., increasing the maximum firing temperature.

4 CONCLUSIONS

This study has taken into account the frost behaviour of 13 industrially-manufactured roofing tiles which were assessed through a double approach: performing severe freeze/thaw testing (EN 539-2) and calculating the durability indices, according to some of the models present in the literature. The products microstructure, in terms of physical, technological and compositional parameters, was fully investigated and the results correlated with the frost resistance of roofing tiles.

Looking at the correlation between the experimental frost behaviour and the calculated durability factors, we can conclude that no model is able to foresee reliably the product performance since the models exhibit a strong dependence on the data population and probably succeed only with a homogeneous sample in terms of both composition and manufacturing technology.

Inserting into a statistical procedure all the microstructural and compositional variables, together with the experimental number of freeze/thaw cycles, new indications came out concerning the design and production of roofing tiles able to withstand adverse climatic conditions. Following these results, it can be pointed out that the production of roofing tiles with excellent frost resistance involves the evaluation of both product (i.e., raw materials composition, microstructure and phase composition of tiles) and processing (i.e., firing temperature) variables.

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