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Recycling PC-TV waste glass in clay bricks and roof tiles

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Abstract. Disposal of PC and TV sets is a growing problem, involving for over 40 wt. % waste glasses with high Pb (funnel) or Ba-Sr concentration (panel) which cannot be recycled in the glass manufacture. A possible way to re-use these glasses is in the manufacturing of clay bricks and roof tiles, that was appraised by laboratory simulation of the brickmaking processing and technological characterization of unfired and fired products. The recycling of both funnel and panel glasses into clay bodies is technologically feasible, resulting in a behaviour substantially plasticity-reducing during shaping-drying (implying a reduction of mechanical strength) and promoting sintering during firing. No significant release of Pb, Ba and Sr was found during firing and leaching test for the carbonate-poor body; in contrast, some Pb volatilization during firing and Sr leaching were observed for the carbonate-rich body. Additions of 2 wt. % appear to be practicable, while 5 wt. % glass induces unacceptable modifications of technological properties. The recommended amount is within 2 and 4 wt. %, depending on the characteristics of clay bodies. The main constraint is that glass must have a particle size below the limit of pan mills used in brickmaking (<1 mm).

1. Introduction

Both technological development and improvement of life standards are bringing about a remarkable growth of the TV-set and personal computer (PC) consumption (Cramer et al., 1995; Menad, 1999; Hermans et al., 2001). Consequently, the amount of wastes from electronic devices is increasing, being for over 40 wt. % represented by cathodic ray tube (funnel) and screen (panel) glasses (Cramer et al., 1995; Menad, 1999). Available estimates are around 25,000 cubic metres per year of PC and TV glass every million people in the European countries (Hermans et al., 2001). Even if the liquid crystals displays are rapidly replacing cathodic tubes, the ‘long wave’ of TV and PC wastes is to be faced on in the forthcoming decade, so that recycling on industrial scale should be the preferable solution; Bernardo et al., 2003; Heart, 2008).

Re-use of PC and TV glasses involves technological problems connected with the high lead concentration (funnel) and the abundance of barium and strontium (panel) (Méar et al., 2006). Recent investigations proposed porcelain stoneware tiles (Rambaldi et al., 2004; Raimondo et al., 2007), ceramic glazes (Andreola et al., 2007), radiation protection tiles (Boccaccini et al., 1997) and glass-alumina platelets composite materials (Minay et al., 2003) as possible applications for recycled PC and TV glasses.

However, these proposals did not find any large-scale development that could represent a viable alternative to waste disposal in landfills. In fact, the economic feasibility of glass recycling needs affordable costs, particularly of waste treatment and transportation, that are difficult to ensure when the waste source is widespread and there is a single end user.

In order to overcome this limit, the chance to have also widespread recycling units is envisaged. This condition is satisfied in the case of brickworks, that in most countries are spread quite uniformly over the territory.

The feasibility of glass recycling into clay bodies was already proved in the case of wastes from car windshields (Mörtel and Fuchs, 1997), soda-lime float and container glasses (Matteucci et al., 2002; Tucci et al., 2004; Pontikes et al., 2004) and glazing lines of tilemaking ceramic industries (Ortelli and Vincenzini, 1983; Palmonari and Tenaglia, 1985; Cava et al., 2000).

This work is aimed at assessing the effect of both funnel and panel glasses on the technological behaviour and technical performance of heavy-clay products. The brickmaking process was simulated at the laboratory scale and the properties of both unfired and fired bricks and roof tiles, containing increasing amount of waste glass, were compared with waste-free reference bodies.

2. Experimental

Two different wastes from the recovery of TV-PC sets (funnel/cathodic tube glass = T and panel/screen glass = S) were selected and their chemical composition, determined by XRF-WDS (Philips, PW 1480), is reported in Table 1. Each glass was dry ground by hammer mill (sieve 0.25 mm).

Two clay bodies were selected as representative of current production of clay bricks (C) and roof tiles (M). These bodies were characterized determining chemical composition (XRF-WDS, Philips, PW 1480), mineralogical composition (XRPD, Rigaku, Miniflex), particle size distribution (X-ray monitoring of gravity sedimentation, Micromeritics, SediGraph 5100). Body C is a carbonate-rich and fine-grained clay, while body M is a relatively coarse-grained clay, poor in carbonates (Table 1).

Five different clay/glass mixtures were experimented for each body: clay without waste (C0 and M0) and with additions of 2 and 5 wt. % funnel glass (CT2, CT5, MT2 and MT5) or panel glass (CS2, CS5, MS2 and MS5).

The technological behaviour of these batches was assessed during body preparation, shaping, drying and firing through a simulation, on a laboratory pilot line, of the industrial processing of clay bricks and roof tiles. In particular, the following working phases were carried out:

(i) clay grinding with a jaw crusher (<20 mm) and a hammer mill (<1 mm);
(ii) hand mixing of clay, glass and water, and successive storage for 7 days;
(iii) plastic extrusion of 100 x 20 x 10 mm^3 bars, with a pneumatic apparatus without vacuum;
(iv) drying at ambient temperature in non-controlled atmosphere for 48 h and then in electric oven at 100 °C overnight;
(v) firing in electric chamber kiln, in static air, at three maximum temperatures (900, 950 and 1000 °C, thermal rate of 100°C/h, 4h soaking) for each series of mixtures.

Unfired and fired products were characterized determining:

- working moisture (ASTM C324, 1992);
- plasticity by means of Pfefferkorn plastic index (Van der Velden, 1979);
- drying and firing shrinkage (ASTM C326, 1997);
- drying behaviour with Adamel apparatus on 80x20x10 mm^3 moulded bars; the drying process is characterized by weight loss occurring in two stages (W1 with shrinkage and W2 without shrinkage) that are graphically represented by the Bigot's curve; a drying

sensitivity index was calculated as (W1·drying shrinkage·0.01)/dry modulus of rupture (Dondi et al., 1998);
- hygroscopicity (Keeling, 1958);
- modulus of rupture of dry and fired samples (ASTM C674, 1994);
- water absorption, open porosity and bulk density (ASTM C373, 1994).

The leaching test (DIN 38414-S4,1984) was performed to check the inertization degree of Pb (glass T) or Ba and Sr (glass S). Samples with addition of 5 wt.% glass were selected: CS5 and CT5 (fired at 950°C) and MS5 and MT5 (fired at 900°C). Bricks were hammer milled to get grains below 2 mm; 12.5 g were put in a 5% (V/V) acetic acid acqueous solution and stirred for 24 hours, keeping pH between 4 and 5. The eluate solutions were analyzed by ICP-OES (Varian, Liberty 200).
On the same samples, the possible volatilization of Pb, Ba and Sr during firing was checked by determining the bulk chemical composition of unfired and fired bodies by means of XRF-EDS (Link-Analytical electron microprobe, 10 analyses each sample).

3. Results and discussion

3.1. Unfired bodies

The addition of waste glass had puzzling effects on technological features of bodies for clay bricks and roof tiles, depending on both composition and granulometric characteristics of clays.

In the body C, both types of glass behave like plasticity-reducing agents as far as drying shrinkage, weight loss with shrinkage and dry bending strength are concerned, especially with the highest waste addition. On the other hand, changes in working moisture and hygroscopicity are negligible (Table 2). Curiously enough, the occurrence of glass seems to enhance plasticity, particularly in the case of funnel, as appreciable by both the Pfefferkorn index (Table 2) and the rheological behaviour calculated from the Pfefferkorn test. In fact, the shear resistance is increased, for the same water content, moving from the waste-free body to those containing panel glass and further funnel glass (Fig. 1). Glass slightly affects the drying behaviour of body C, as it can be seen from Bigot’s curves (Fig. 2).

On the other hand, waste glasses influence in a controversial way the body M. Funnel glass additions increase working moisture and drying shrinkage, while panel glass decreases the body contraction and does not affect mechanical strength when added at 2 wt. % (Table 2).

Overall, drying sensitivity is increased in all glass-bearing bodies, essentially for their lowered mechanical resistance, which appears to be the main limitation of this waste recycling (Brown and Mackenzie, 1982).

3.2. Fired bodies

Waste glasses have a variable effect on the physical properties of fired bricks, depending on the body (type of glass and clay) and firing conditions (Tables 3 and 4).

Generally speaking, glass tends to increase firing shrinkage and to a certain extent to improve bending strength, due to lower values of water absorption and higher values of bulk density found in glass-bearing bricks. Such a general picture is expected for low-melting glasses, which basically play the role of sintering promoters through the formation of a viscous phase at high temperature (Matteucci et al., 2002; Tucci et al., 2004). Similar behaviours have been already observed in clay bodies containing glass as raw material

(Ortelli and Vincenzini, 1983; Palmonari and Tenaglia, 1985; Cava et al., 2000; Matteucci et al., 2002; Pontikes et al., 2004; Tucci et al., 2004).

Looking in detail, differences arise between bodies M and C, as between funnel and panel glasses. In the body M, whose proper firing temperature is close to 900 °C, the panel glass is easily incorporated in the clay matrix without significant changes of physical properties. Increasing temperature to 1000 °C implies an enhanced sintering degree: lower water absorption, higher firing shrinkage and bulk density, better mechanical performance. Analogous trends are shown by the body M containing funnel glass, even if firing at 900 °C is not fully satisfactory, as both bulk density and bending strength are decreased with respect of the waste-free body.

In the clay C, there are limited changes in the physical properties over the firing range: for both glasses, even with 5 wt. % addition, the values of technological properties are close to those of the reference body in between 950 and 1000 °C.

The stability during firing of elements characteristic of waste glasses was checked by comparing the Pb, Ba and Sr concentrations in unfired and fired bodies (Fig. 3). The points falling below the straight line 1:1 imply a volatilization, particularly of lead in the body C, which has a higher optimal firing temperature (950 °C versus 900 °C of body M). Lead volatilization from glazes is known in the ceramic tile industry (Dondi et al., 1990). No clue of Pb, Ba or Sr mobilization was found for the body M.

The inertization degree of waste glasses was verified through a leaching test (Table 5). The lead concentration in the eluates is always below the detection limit, implying a full stabilization, which confirms previous data on the same glass put into a different type of ceramic body (Raimondo et al., 2007). Very low amounts of barium were released by both C and M bodies (2-3 mg/kg); a similar figure was measured for Sr leached from the body M (7 mg/kg). These values are in the order of one thousandth of the total concentration in the body (Table 5). A different case is that of Sr leached from the body C (about 60 mg/kg); in this circumstance, an important contribution from the body may be invoked, since it contains a significant fraction of Sr-rich calcareous microfossils, which is likely to be easily leachable even after firing.

3.3. Recycling perspectives

In Italy, the expected amount of PC-TV glasses to be recycled may be estimated around 50,000 tpy (tons per year), assuming a mean value of 0.7 TV-PC sets for every inhabitant, an average lifetime of 7.5 years, and 9 kg of glass for each set, i.e. 59 million inhabitants x 0.7 sets x 0.133 lifetime x 0.009 tons (Menad, 1999).

Taking into account that the mean output of a brickmaking plant in Italy is around 100,000 tpy, the capacity of a single plant to recycle PC-TV glass – considering a prudential threshold of 2-3 wt. % – would be from 2,000 to 3,000 tpy. However, not every clay body is plastic enough to afford the addition of a plasticity-reducing materials like glass. Therefore, the estimated 50,000 tpy of waste glass could be absorbed in the production by 20 to 30 brickworks out of the about 160 currently operating in the country.

4. Conclusions

Recycling of PC-TV funnel and panel glasses into clay brick and roof tile bodies is technologically feasible. The main constraint is to have ground glass with a particle size below the limit of pan mills used in brickmaking (i.e. below 0.8 to 1 mm). A word of caution must be spent in the case of carbonate-rich bodies, for which some lead volatilization during firing and some strontium leaching were observed.

Additions up to 2 wt. % of glass to the clay bodies do not bring about significant changes of technological performances of both unfired and fired products. In contrast, additions of 5 wt. % glass may have deleterious effects, particularly on the mechanical strength, depending on the glass type and the characteristics of the clay body.

The expected amount of waste glass from PC-TV dismantling in Italy, i.e. around 50,000 tpy, could be recycled by 20 to 30 brickworks adding 2 wt. % of ground glass to the clay body.

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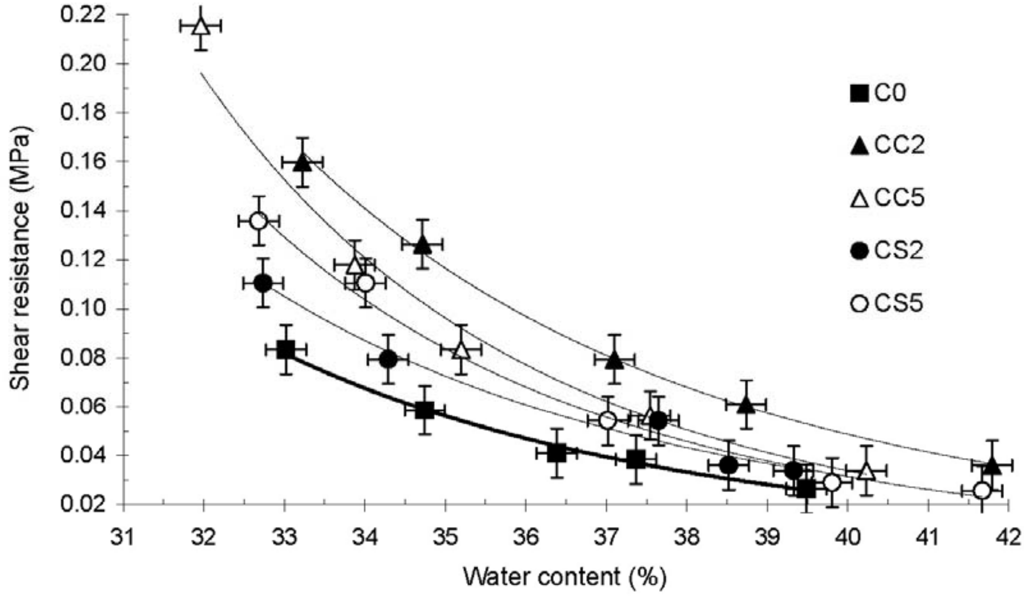


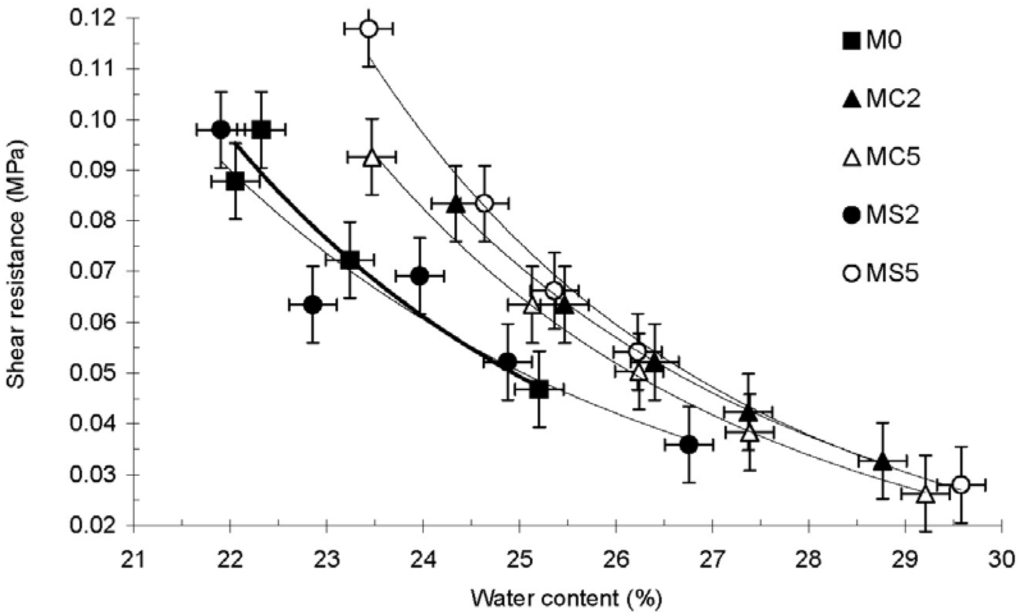
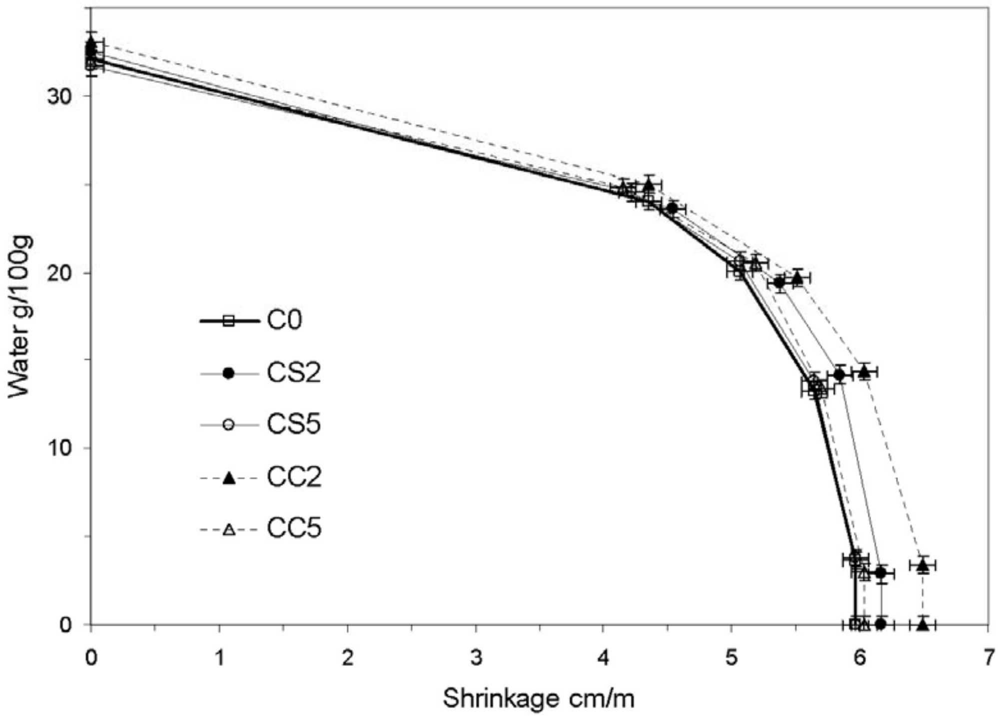


Fig. 1. Rheological behaviour of extruded bodies estimated by Pfefferkorn test: shear resistance in function of water content for roof tile bodies (M) and clay brick bodies (C) added with PC-TV (C: cathodic tube, S: screen) waste glass.



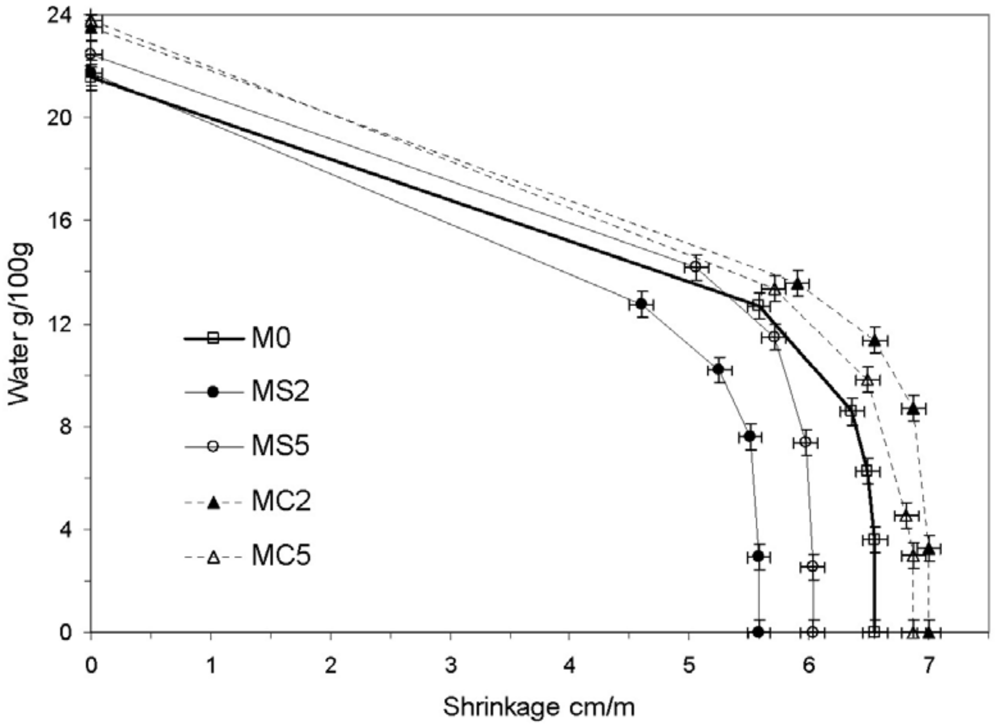


Fig. 2. Drying behaviour of extruded bodies estimated by Bigot's curves for clay brick bodies (C) and roof tile bodies (M) added with PC-TV (T: cathodic tube, S: screen) waste glass.

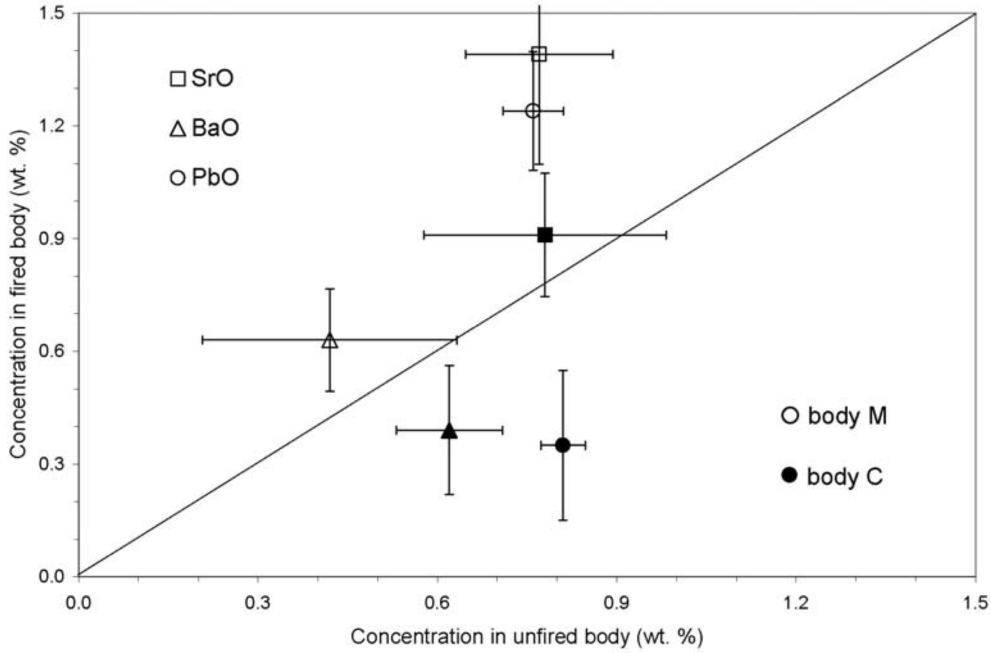


Fig. 3. Estimation of element volatilization during firing of glass-bearing bricks (CS5, CT5, MS5, MT5): concentration of PbO, BaO and SrO in the unfired body versus the fired body (XRF-EDS, values corrected for the loss on ignition).

Table 1
Chemical and mineralogical composition and particle size distribution of the bodies for clay bricks (C) and roof tiles (M) and PC-TV waste glasses (T: cathodic tube, S: screen).

	wt.%	C	M	T	S
Chemical composition	SiO_2	43.8	56.6	51.6	62.7
	TiO_2	0.6	0.7	0.1	0.4
	ZrO_2	<0.1	<0.1	0.2	1.9
	Al_2O_3	12.8	17.3	3.6	2.3
	Fe_2O_3	4.5	6.9	0.1	0.1
	MgO	2.8	3.0	1.9	0.3
	CaO	15.2	3.3	3.8	1.0
	SrO	<0.1	<0.1	0.7	7.6
	BaO	<0.1	<0.1	0.8	8.4
	PbO	<0.1	<0.1	23.1	0.4
	Na_2O	0.8	0.8	6.1	7.4
	K_2O	2.0	2.1	7.5	7.1
	L.o.I.	16.5	8.7	<0.1	<0.1
Mineralogical composition	Illite	21	23		
	Chlorite	5	11		
	Kaolinite	13	9		
	Smectite	<1	13		
	Quartz	17	27		
	Plagioclase	7	4		
	K-Feldspar	3	0		
	Calcite	26	6		
	Dolomite	2	<1		
	Fe-oxyhydroxides	5	5		
	Accessories	1	2		
Particle fractions	> 63 µm	4	3		
	> 20 µm	13	13		
	> 10 µm	24	29		
	> 2 µm	55	69		

Table 2
Technological behaviour of the unfired bodies for clay bricks (C0) or roof tiles (M0) containing screen glass (CS2, CS5, MS2, MS5) or cathodic tube glass (CT2, CT5, MT2, MT5).

Parameter	Unit	C0		CS2		CS5		CT2		CT5	
		mean	s.d.	mean	s.d.	mean	s.d.	mean	s.d.	mean	s.d.
Working moisture	wt.%	33.3	0.1	33.7	0.1	33.4	0.1	33.9	0.1	32.3	0.1
Pfefferkorn index	wt.%	43.9	0.9	44.8	6.0	42.2	2.4	50.6	5.0	46.8	3.9
Drying shrinkage	cm/m	6.8	0.1	6.6	0.5	6.3	0.3	6.5	0.3	6.3	0.1
Weight loss with shrinkage	%	35	1	37	1	32	1	37	1	32	1
Drying sensitivity	1	0.37	0.01	0.44	0.01	0.46	0.01	0.37	0.01	0.43	0.01
Hygroscopicity	wt.%	1.57	0.11	1.52	0.13	1.72	0.16	1.61	0.13	1.50	0.19
Dry bending strength	MPa	6.4	0.3	5.5	0.6	4.3	0.1	6.4	0.3	4.7	0.1
Parameter	Unit	M0		MS2		MS5		MT2		MT5	
		mean	s.d.	mean	s.d.	mean	s.d.	mean	s.d.	mean	s.d.
Working moisture	wt.%	23.4	0.1	23.1	0.1	23.6	0.1	24.9	0.1	24.8	0.1
Pfefferkorn index	wt.%	40.6	2.3	32.4	3.5	31.3	1.5	32.6	1.2	30.3	3.0
Drying shrinkage	cm/m	6.1	0.4	6.1	0.1	6.5	0.1	6.8	0.2	6.6	0.3
Weight loss with shrinkage	%	48	1	50	1	44	1	50	1	53	1
Drying sensitivity	1	0.33	0.01	0.34	0.01	0.40	0.01	0.50	0.01	0.50	0.01
Hygroscopicity	wt.%	1.94	0.11	1.88	0.04	1.64	0.18	2.01	0.19	1.57	0.09
Dry bending strength	MPa	8.9	0.6	9.1	0.7	7.0	0.4	6.8	0.9	7.0	0.3

Table 3
Technological properties of the body for clay bricks (C0) containing 2% or 5% screen glass (CS2 and CS5) or cathodic tube glass (CT2 and CT5) fired at maximum temperature of 900, 950 and 1000 °C.

	Parameter	Unit	C0		CS2		CS5		CT2		CT5	
			mean	s.d.	mean	s.d.	mean	s.d.	mean	s.d.	mean	s.d.
900 °C	Firing shrinkage	cm/m	2.8	*0.2*	3.0	*0.5*	3.5	*0.2*	3.2	*0.3*	3.2	*0.4*
	Bending strength	MPa	19.8	*2.1*	18.2	*0.7*	17.9	*2.0*	18.1	*3.8*	20.1	*3.4*
	Water absorption	wt.%	17.8	*0.4*	18.7	*0.2*	18.5	*0.6*	18.4	*0.6*	17.2	*1.2*
	Open porosity	% vol.	30.4	*0.6*	31.5	*0.3*	31.3	*0.7*	31.2	*0.7*	29.7	*1.5*
	Bulk density	g/cm^3	1.705	*0.011*	1.685	*0.008*	1.694	*0.015*	1.699	*0.024*	1.734	*0.031*
950 °C	Firing shrinkage	cm/m	2.8	*0.2*	2.8	*0.3*	3.2	*0.3*	3.1	*0.4*	2.9	*0.3*
	Bending strength	MPa	19.8	*0.8*	19.6	*1.2*	22.8	*1.8*	17.3	*3.2*	18.3	*1.8*
	Water absorption	wt.%	18.1	*0.7*	18.2	*0.3*	17.9	*1.1*	18.9	*0.2*	17.8	*0.8*
	Open porosity	% vol.	31.0	*1.0*	31.4	*0.6*	30.6	*1.4*	31.9	*0.3*	30.6	*0.9*
	Bulk density	g/cm^3	1.706	*0.017*	1.723	*0.037*	1.716	*0.036*	1.692	*0.006*	1.721	*0.025*
1000 °C	Firing shrinkage	cm/m	2.6	*0.2*	3.2	*0.7*	3.1	*0.2*	3.2	*0.6*	3.0	*0.5*
	Bending strength	MPa	21.8	*2.1*	21.1	*0.2*	22.6	*2.7*	18.5	*1.7*	18.1	*1.9*
	Water absorption	wt.%	18.2	*0.5*	17.8	*0.5*	17.4	*0.9*	18.3	*0.3*	17.3	*0.1*
	Open porosity	% vol.	30.1	*0.7*	29.8	*0.6*	29.6	*1.0*	30.5	*0.3*	29.2	*0.2*
	Bulk density	g/cm^3	1.653	*0.013*	1.675	*0.013*	1.701	*0.035*	1.665	*0.012*	1.690	*0.005*

Table 4
Technological properties of the body for roof tiles (M0) containing 2% or 5% screen glass (MS2 and MS5) or cathodic tube glass (MT2 and MT5) fired at maximum temperature of 900, 950 and 1000 °C.

	Parameter	Unit	M0		MS2		MS5		MT2		MT5	
			mean	s.d.	mean	s.d.	mean	s.d.	mean	s.d.	mean	s.d.
900 °C	Firing shrinkage	cm/m	1.3	0.3	1.6	0.2	1.3	0.2	1.8	0.3	2.1	0.1
	Bending strength	MPa	17.0	0.4	17.8	0.4	17.0	1.1	16.3	1.5	15.9	0.8
	Water absorption	wt.%	8.7	0.1	8.7	0.1	8.9	0.2	8.8	0.2	8.4	0.1
	Open porosity	% vol.	17.6	0.2	17.5	0.1	17.8	0.4	17.6	0.4	16.8	0.1
	Bulk density	g/cm^3	2.017	0.004	2.017	0.003	2.000	0.009	2.008	0.009	2.014	0.004
950 °C	Firing shrinkage	cm/m	2.0	0.4	2.5	0.2	2.4	0.4	2.5	0.2	3.0	0.3
	Bending strength	MPa	16.4	1.0	17.1	0.9	16.7	0.8	16.9	0.7	16.7	0.7
	Water absorption	wt.%	7.3	0.1	7.3	0.1	7.3	0.2	7.2	0.1	6.8	0.2
	Open porosity	% vol.	15.1	0.2	14.9	0.1	15.0	0.3	14.8	0.2	14.1	0.4
	Bulk density	g/cm^3	2.061	0.012	2.058	0.010	2.055	0.011	2.062	0.009	2.078	0.012
1000 °C	Firing shrinkage	cm/m	3.1	0.2	3.5	0.3	3.6	0.2	3.6	0.4	4.0	0.3
	Bending strength	MPa	20.3	0.4	21.5	1.5	20.9	0.7	21.3	1.2	20.0	1.7
	Water absorption	wt.%	6.0	0.1	5.6	0.1	5.7	0.3	5.6	0.1	5.3	0.1
	Open porosity	% vol.	12.4	0.2	11.7	0.2	11.9	0.5	11.7	0.2	11.1	0.3
	Bulk density	g/cm^3	2.067	0.005	2.074	0.007	2.068	0.025	2.083	0.008	2.097	0.002

Table 5
Concentration of Pb, Ba and Sr in the bodies C (samples CT5 and CS5) and M (samples MT5 and MS5):
total amount in the fired body and leachable fraction (DIN 38414-4).

Element	Body C		Body M	
	Total (mg/kg)	Leachable (mg/kg)	Total (mg/kg)	Leachable (mg/kg)
Pb	3,700	<1	12,100	<1
Ba	4,000	2 ± 1	5,800	3 ± 1
Sr	12,200	60 ± 5	8,800	7 ± 1