

CERAMIC TILES OBTAINED FROM PACKING GLASS CULLETS

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ABSTRACT

Powder compaction is a popular route for the production of components followed by sintering in a hot state. Densification occurs by sliding and restacking of powder particles at low pressures, which leads to rearrangement of the powder from a loose array to close packing. In this work the compaction behaviour of glass particles was studied. Packing glass cullets were milled and pressed in order to obtain ceramic tiles. A factorial 3^k design approach was used to study the influence of milling time, binder and plasticizer addition and pressing pressure in the apparent density and fracture loading of the tiles. The results show the influence of the particle size distribution on the compaction behaviour of the glass tiles.

1. INTRODUCTION

Powder compaction is a popular route for the production of components followed by sintering in a hot state. Generally most of the densification takes place in the cold consolidation step^[1-2]. On a fundamental level, densification occurs simply by the motion of particle centres toward each other in several stages. The first stage of the compaction process is sliding and restacking of powder particles at low pressures, which leads to rearrangement of the powder from a loose array to close packing. At a certain relative density, the particles are more or less in a fixed arrangement. Due to the resistance of a material against deformation, the internal stress of the particles increases. If the applied stress is released before a critical state of deformation, the particles deform elastically, i.e. the deformation is reversible, and the particles inside the powder bed regain their original shapes. In the following stage, for brittle systems, particles fragment into smaller units at fracture strength, σ_f . Material fracture will eventually occur at higher deformations. Lower compressibility results when powder particles are strain hardened during disintegration^[1, 3-6].

There are so many effective parameters in compressibility behaviours of materials, such as intrinsic characteristics of the material, deformability, morphology, particle shape, Interparticle and particle/die wall friction^[10], internal porosity, particle size distribution, pressure applying for consolidation, etc. With increasing applied pressure, the density of the powder mass increases or porosity decreases. The determination of the densification behaviour of a powder is usually based on the measurement of the relative density of the powder bed as a function of the compaction pressure. However there are no fully acceptable expressions to describe accurately the extent of densification versus applied pressure for powders, although many compaction equations have been suggested^[1, 7-9].

Compaction equations usually were used to develop a linear relationship between pressure and relative density. So it would be possible to make comparisons between series of data and/or prediction a required pressure to obtain a certain level of density. Some researchers have tried to correlate these parameters with mathematical equations to determine the effect of mechanical milling on morphology of powders and have related the compaction behaviour of powders to the microstructural evolutions occurred during milling^[1, 3-7].

The addition of organic binders in granulated ceramic powders increases markedly the mechanical resistance of green product. When the binder is ductile (glass transition temperature lower than forming temperature), the mechanical resistance increases rapidly and linearly with forming pressure until it reaches a plateau above a transition pressure. The major contribution of the binder to the mechanical resistance is attributed to the binder located inside the granules with a low contribution of the organic-rich shell around the granules^[10].

The evolution of the mechanical resistance probably results from an increase in the contact surface of the polymer bridges between primary ceramic particles considered covered by a binder film. The estimation of the fracture energy of the pressed green compacts suggests that a plastic deformation takes place during failure. When the glass transition temperature of the binder is higher than the pressing temperature, microcracking of the brittle polymer-rich shells surrounding the granules seems to be responsible for the low values of mechanical resistance

until the forming pressure become large enough to close these microcracks through densification of the whole assembly. Then, the mechanical resistance increases rapidly^[10].

In this work the compaction behaviour of glass particles is studied. Packing glass cullets were milled and pressed in order to obtain ceramic tiles. A factorial design approach was used to study the influence of milling time, binder and plasticizer addition and pressing pressure.

2. MATERIALS AND METHODS

Packing glass cullets were collected, washed and fragmented. A 3^{k-1} factorial design was used to determine the compaction behaviour of the glass cullet. Milling time (8h, 12h and 16h), additive content (0.25%/0.25%, 0.50%/0.50% and 0.75%/0.75%) and pressing pressure (300kgf/cm², 400kgf/cm² and 500kgf/cm²) were used as main factors resulting in a fractioned factorial design with nine runs. The fragmented cullets were milled with water in a laboratory jar mill according the design milling times and additive content. After milling the slurries were dried (110°C, 24h) and the powder granulated with 6% water. Polyvinyl alcohol was used as binder and acrylic resin was used as plasticizer. The granulated powder was compacted in a laboratory press according the design into 20mm x 120mm compacts. Finally, the apparent density (immersion method) and the fracture at maximum loading (ISO 10545) of the compacts were determined for all nine runs. The results were analyzed by analysis of variance (ANOVA) and plotted as response surfaces.

3. RESULTS AND DISCUSSION

Table 1 shows the results for apparent density and fracture at maximum loading for the 3^{k-1} factorial design.

RUN	MILLING TIME (h)	ADDITIVE (%)	COMPACTION (kgf/cm ²)	DENSITY (g/cm ³)	RUPTURE (kgf)
1	8	0.25/0.25	300	1.360±0.015	2.35±0.57
2	8	0.50/0.50	500	1.400±0.019	2.85±0.36
3	8	0.75/0.75	400	1.300±0.032	2.29±0.30
4	12	0.25/0.25	500	1.403±0.050	1.49±0.19
5	12	0.50/0.50	400	1.232±0.024	1.84±0.51
6	12	0.75/0.75	300	1.169±0.008	0.86±0.19
7	16	0.25/0.25	400	1.155±0.018	1.37±0.28
8	16	0.50/0.50	300	1.069±0.010	1.57±0.15
9	16	0.75/0.75	500	1.109±0.007	1.75±0.28

Table 1. Apparent density (g/cm³) and rupture at maximum loading (kgf) in function of milling time (h), additive content (%) and pressure of compaction (kgf/cm²)

The results for apparent density were analyzed by ANOVA, table 2. The major effect is caused by the milling time at a high significance level (99%). The factor interactions were not considered because the design is fractioned. Figure 1 shows the response surface graphs for the apparent density.

APPARENT DENSITY (g/cm ³)	SS	dF	MS	F	p
milling time	0.090656	2	0.045328	103.6203	0.009558
additive content	0.019758	2	0.009879	22.5829	0.042404
pressure of compaction	0.017460	2	0.008730	19.9571	0.047717
total SS error	0.000875	2	0.000437		
total SS	0.128749	8			

Table 2. ANOVA for apparent density in function of milling time (h), additive content (%) and pressure of compaction (kgf/cm²)

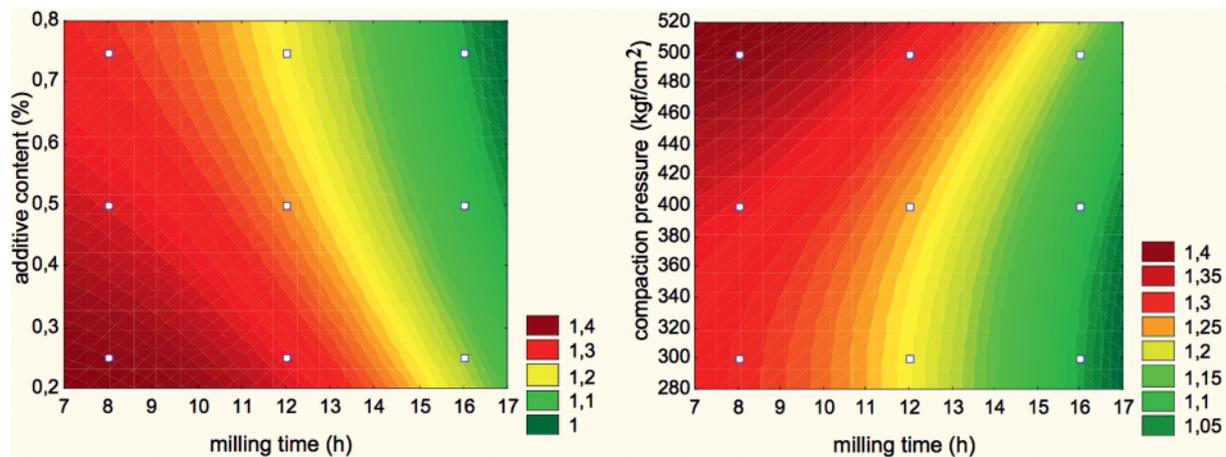


Figure 1. Response surface for the apparent density of the compacted glasses

The apparent density is strongly affected by the milling time. The combination of 8h milling time with high compaction pressure (500kgf/cm²) and a small amount of additives (0.25% PVA and 0.25% acrylic resin) results in the best system packing. The model fit is very high (R²=0.993), validating the analysis.

Table 3 shows the analysis of variance (ANOVA) for the rupture at maximum loading. Once more the major effect is caused by the milling time at a high significance level (94%). The factor interactions were not considered because the design is fractionated. Figure 2 shows the response surface graphs for the rupture at maximum loading.

RUPTURE AT MAXIMUM LOADING (kgf)	SS	dF	MS	F	p
milling time	2.108889	2	1.054444	16.35361	0.057625
additive content	0.338689	2	0.169344	2.62640	0.275756
compaction pressure	0.286956	2	0.143478	2.22523	0.310056
total SS error	0.128956	2	0.064478		
total SS	2.863489	8			

Table 3. ANOVA for rupture at maximum loading in function of milling time (h), additive content (%) and pressure of compaction (kgf/cm²)

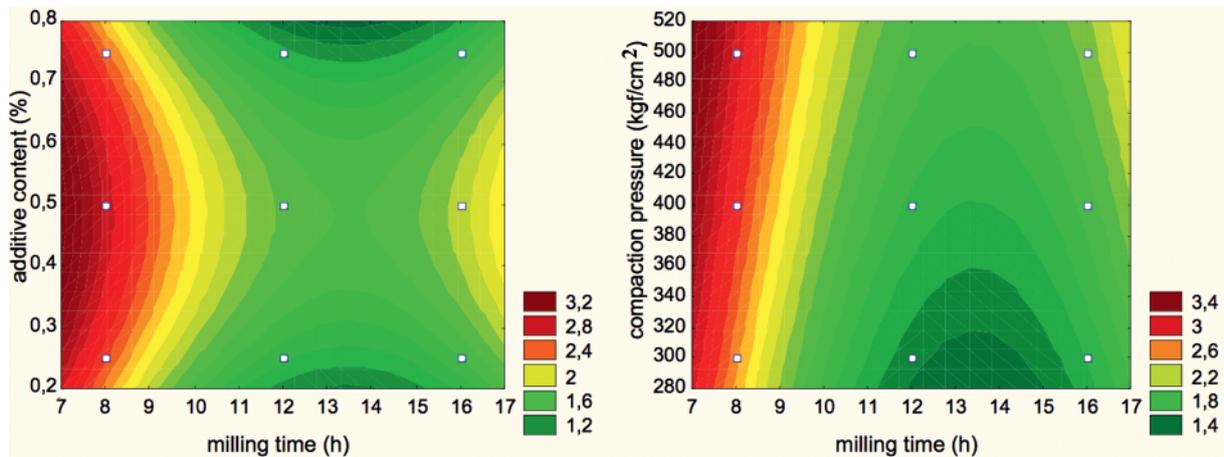


Figure 2. Response surface for the rupture at maximum loading of the compacted glasses

The rupture at maximum loading is also strongly affected by the milling time. The combination of 8h milling time with high compaction pressure (500kgf/cm²) and a medium amount of additives (0.50% PVA and 0.50% acrylic resin) results in the best system packing. The model fit is very high ($R^2=0.955$), validating the analysis.

Finally, figure 3 shows the particle morphology after milling. The 8h milling time presents a large particle size distribution, with particles ranging from 10 μ m to 30 μ m. The 16h milling time shows a narrow particle size distribution, with particles ranging from 5 μ m to 15 μ m. The particle size distribution was not carried out, so this analysis is only qualitative based on the micrographs.

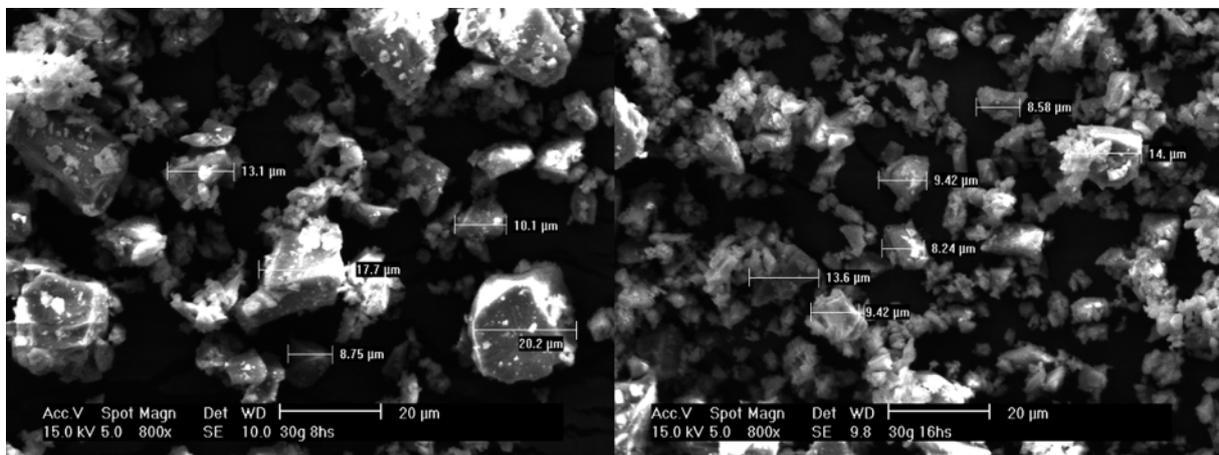


Figure 3. Particle morphology after 8h and 16h milling times

4. CONCLUSION

The recycling of glass cullet in ceramic tiles is an easy and efficient way to reduce the disposal of this kind of residue. Because the glass cullet is the only raw material and because the cullet can be processed in a regular ceramic tile plant, without any kind of adaptation, the solution could be rapidly adopted to transform the vitreous residues of municipal incinerator plants into ceramic tiles. The process is green, because it uses residues and do not create any residue at all. Only water and small

fractions of organic additives are used in the process, resulting in products with a large and well established market.

Regarding the results obtained in this preliminary work, the apparent density and the fracture at loading are strongly affected by the milling time. The combination of a reduced milling time with high compaction pressures and a small to medium amount of additives results in the best system packing. The particle size distribution of the glass residue is the most important factor to consider in the compaction of non plastic brittle materials. The next step is the study of the sintering behaviour of the compacted glass system.

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