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# 1 Waste recycling in ceramic tiles: a technological outlook

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## 4 Abstract

5 The ceramic industry is going to be deeply involved in the transition to a circular economy. However,  
6 the main obstacle to a widespread recourse to waste recycling is the lack of knowledge about its  
7 effect in ceramic tile manufacturing. The rationale behind this work is to look at recycling from the  
8 industrialist's point of view. The effects of different kinds of waste on technological behavior,  
9 technical performance, and environmental impact of ceramic tiles are discussed in the prospect of  
10 an industrial transfer. Technical constraints that waste recycling must comply to be transferable to  
11 ceramic tile manufacturing are examined in detail. Available information on the behavior of ceramic  
12 tile bodies containing wastes from various sources is reviewed to get an outlook about feasibility  
13 (expressed in terms of Technology Readiness Level) and recommended recyclable amount. In  
14 addition, hindrances to scale up and matters of concern (e.g., hazardous components and gaseous  
15 emissions) are pointed out and discussed.

16 *Key-words:* ceramic tile; technical performance; technological behavior; waste recycling.

## 17 1. Introduction

18 Waste recycling in ceramic tiles can be traced back to the pioneering study of [Kats and](#)  
19 [Kvyatkovskaya \(1972\)](#) and a few others issued in the 1980s and early 1990s ([Higgins et al., 1979;](#)  
20 [Brown and Mackenzie, 1982;](#) [Abdrakhimov et al., 1990;](#) [Dondi et al., 1990, 1992](#)). In the same period,  
21 the ceramic industry began to recycle its own processing residues and a manifesto of this early  
22 cannibalistic view appeared in 1991 ([Manfredini et al., 1991](#)). Nonetheless, the academic and  
23 industrial interest was very limited until 2000; since then, scientific papers appeared at a growing  
24 rate: in the last decade over 20 publications per year. On the industrial side, even before the current  
25 traction towards a circular economy, waste recycling was mainly driven by marketing reasons,  
26 supported through green certificates, like the LEED credits ([Palmonari and Timellini, 2000;](#) [Blengini](#)  
27 [and Shields, 2010;](#) [Gabaldón-Estevan et al., 2014](#)), and the incoming standard on the sustainability  
28 of ceramic tiles ([ISO/DIS 17889, 2019](#)).

29 The main obstacle to a widespread recourse to waste recycling is the lack of knowledge about the  
30 effect of residues in ceramic tile manufacturing. This is a multi-sided issue that has various aspects  
31 to consider, in particular:

32 *Technological behavior*, i.e. the effect of waste addition on the way bodies behave along the  
33 manufacturing line (that must be within the variance tolerated in the industrial practice).

34 *Technical performance*, i.e. the repercussion of waste introduction on the properties of finished  
35 products and the conformity to standard specifications and market requirements.

36 *Environmental impact*, seen as the way the waste presence in ceramic bodies affects the solid  
37 (scraps), liquid (wastewater, leachates) and gaseous emissions (flue gas from kilns).

38 *Economic sustainability*, accounting for the balance between cost and benefit (in terms of energy  
39 consumption, investments, transportation, and so on).

40 The goal of the present paper is to overview the effect of waste recycling on technological behavior  
41 and technical performance of ceramic tiles, with some concern on the environmental impact.  
42 However, any life cycle assessment of waste addition in tile-making is not considered here. The  
43 rationale behind the analysis of the literature is directed towards two outcomes. First, an overall  
44 examination of technical constraints that waste recycling must comply to be transferable to ceramic  
45 tile manufacturing. Second, a detailed critical review of available information on the technological  
46 behavior of ceramic tile bodies containing wastes from various sources. Finally, a technological  
47 outlook regards feasibility, recyclable amount, and possible matters of concern.

## 48 **2. General issues of waste recycling in ceramic tile-making**

49 This section presents the modern ceramic tile technology, focusing on issues relevant for waste  
50 recycling. This introductory part offers an industry-oriented view on the target of waste incorporation  
51 into ceramic tile bodies and a general review of potential effects of waste occurrence in the various  
52 production stages.

### 53 *2.1. Constraints from ceramic tile manufacturing*

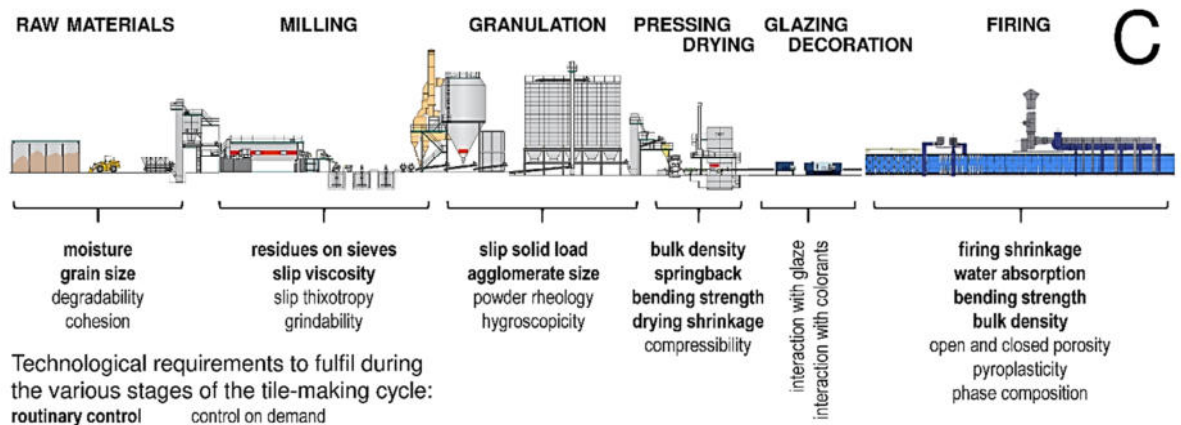
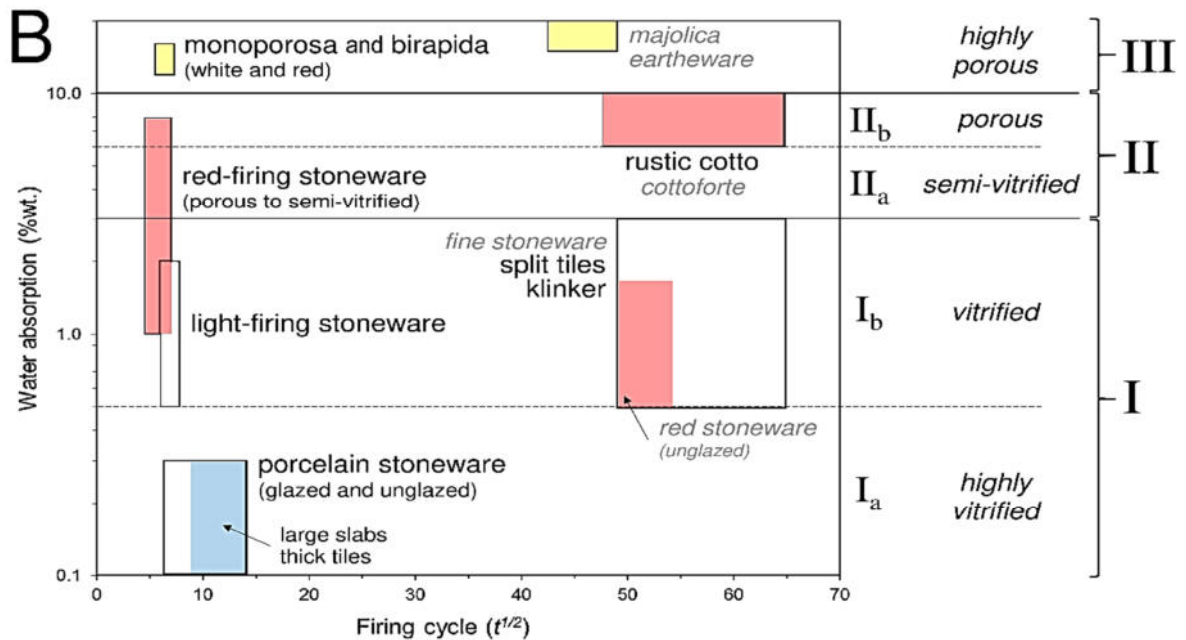
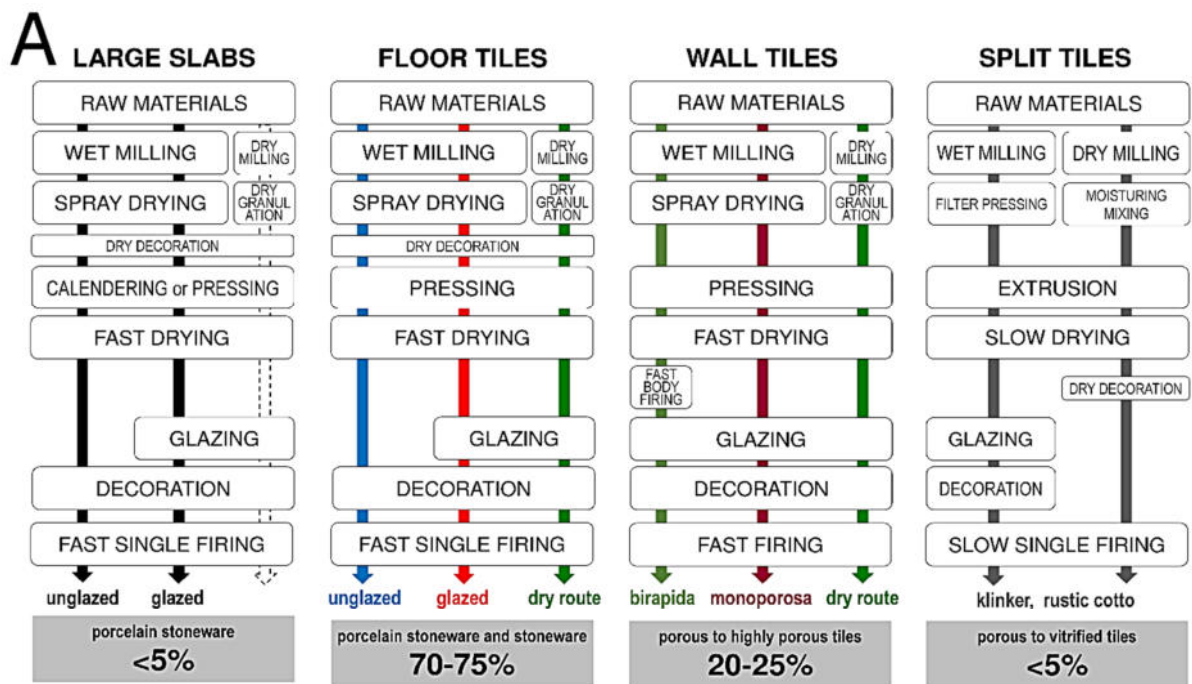
54 Although all ceramic tiles have been manufacturing with the classical scheme (milling → granulation  
55 → shaping → drying → glazing → decoration → firing) there are several variants developed to

56 produce different typologies, in the framework of a continuous innovation (Dondi et al., 2014;  
57 Gabaldón-Estevan, 2016).

58 Ceramic tiles have distinct product specifications according to end-use: walling and flooring (indoor  
59 or outdoor) plus new applications that recently gained different markets (interior design, etc). On the  
60 other hand, the classification of ceramic tiles is based on water absorption and shaping technique,  
61 defining three main classes (I, II, III) with subclasses (e.g., I<sub>a</sub> and I<sub>b</sub>) declined for extruded (prefix A)  
62 or pressed (prefix B) products. This matrix gives rise to twelve groups, each having its own  
63 specifications, mandatory or voluntary (ISO 13006, 2018). For sake of simplicity, we will refer to five  
64 categories of ceramic bodies with increasing water absorption (WA): highly vitrified (group BI<sub>a</sub>,  
65 WA<0.5%); vitrified (BI<sub>b</sub> or AI<sub>b</sub>, 0.5%<WA<3%); semi-vitrified (BII<sub>a</sub>, 3%<WA<6%); porous (BII<sub>b</sub> or  
66 All<sub>b</sub>, 6%<WA<10%); highly porous (BIII, WA<10%). In order to satisfy such different standard  
67 specifications, the ceramic tile industry developed various product typologies, each featured by its  
68 own combination of batch composition, manufacturing technology, and market destination. Every  
69 typology took its own commercial name that does not match the standard terminology, so giving rise  
70 to a sort of “double nomenclature”, which could create some confusion (see Table 1).

71 Table 1. Nomenclature, production technology, end-use, classification, and main standard  
72 requirements for the more common typologies of ceramic tiles currently on the market.

Commercial name	Technology (see Fig. 1A)	Major end-use	Classification ISO13006 standard	Water absorption (% wt.)	Modulus of rupture (MPa)	Deep abrasion (mm <sup>3</sup> )
Porcelain stoneware	Large slabs	ventilated façades interior design	BI <sub>a</sub> (Group G)	<0.5	>35	<175
	Floor tiles	flooring (indoor & outdoor) walling (outdoor)	BI <sub>a</sub> (Group G)	<0.5	>35	<175
Light-firing stoneware	Floor tiles	flooring (indoor)	BI <sub>b</sub> (Group H)	0.5-3	>30	<175
Red-firing stoneware	Floor tiles	flooring (indoor)	BI <sub>b</sub> -BII <sub>a</sub> (Groups H-J)	0.5-6	>30 >22	<175 <345
	Wall tiles	walling (indoor)	BII <sub>b</sub> (Group K)	6-10	>18	<540
Monoporosa Birapida	Wall tiles	walling (indoor)	BIII (Group L)	>10	>15	NA
Klinker	Split tiles	flooring (indoor & outdoor)	AI <sub>b</sub> (Group A)	0.5-3	>23	<275
Rustic cotto	Split tiles	flooring (indoor & outdoor)	All <sub>b</sub> (Groups D-E )	6-10	>9 >17.5	<1062 <649



73  
74 Figure 1. Flowchart of the main technologies used in ceramic tile manufacturing (A). Classification of various  
75 typologies of ceramic tiles versus duration of firing cycle (B). Quality controls in the distinct stages of ceramic  
76 tile manufacturing (C).

77 The manufacturing solutions currently in use can be summarized into four main technologies: floor  
78 tiles and wall tiles (both pressed); split tiles (extruded); large slabs obtained through novel  
79 compaction techniques (Fig. 1A).

80 Although they present many stages in common, it must be considered that actual manufacturing  
81 conditions are completely distinct for each typology (e.g., body composition, particle size distribution,  
82 specific pressure of compaction, firing schedule). The diffusion of these technologies can be  
83 estimated as follows: floor tiles, 70-75% of the global output; wall tiles, 20-25%; the others occupy a  
84 market niche (each <5%): one fading out (split tiles) and the other growing up (large slabs).

85 Such manufacturing conditions – specific for every technology – are of paramount importance for  
86 making waste recycling feasible. They must be carefully considered when the waste addition is  
87 designed and experimented. The choice of a given set of working conditions implicitly addresses the  
88 conclusions to a determined technology, as often occurs in the literature, beyond what is the target  
89 declared by authors. This is particularly true about the firing schedule since every tile typology has  
90 its own combination of water absorption and firing conditions (Fig. 1B).

91 Any waste-bearing body must comply all technological requirements along the manufacturing line  
92 as well as all mandatory technical properties for the finished product. In the industrial practice, the  
93 technological behavior is kept under control in every stage of the production line: the basic properties  
94 measured on semi-finished products are listed in Figure 1C. In addition, the most important technical  
95 properties of finished products are summarized in Table 1.

## 96 *2.2. Target of waste incorporation in ceramic tiles*

97 Wastes are intended to turn into secondary raw materials and to substitute – partially or entirely –  
98 naturally-occurring industrial minerals. This implies that wastes must comply a set of requirements  
99 that concerns any raw material for ceramic tiles, as will be seen in more detail in section 2.3.

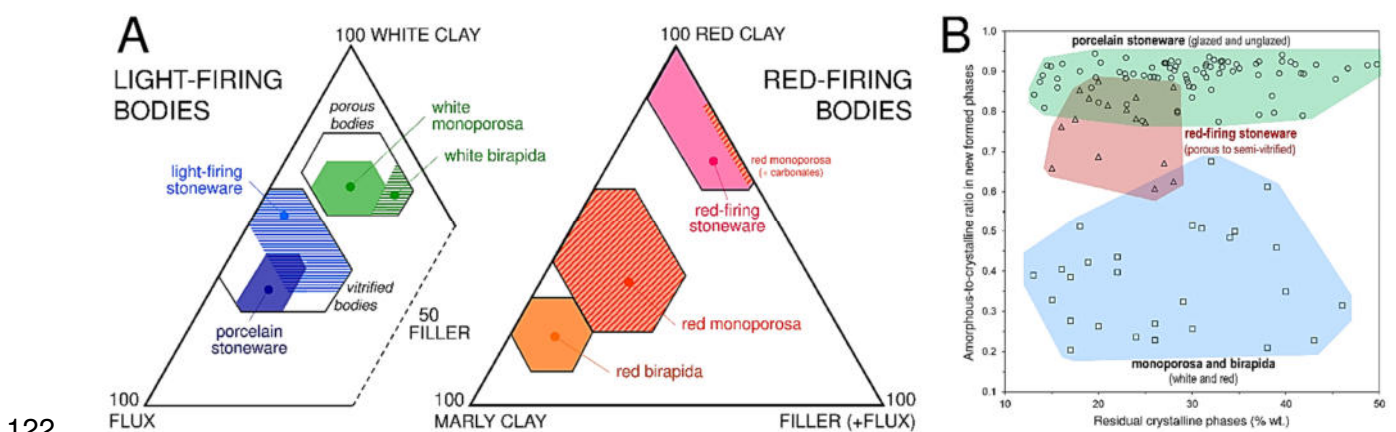
100 Account must be taken that ceramic raw materials play the following roles in the batch (Dondi et al.,  
101 2014; Dondi, 2018):

- 102 • *Plasticity provider* helps forming a stable slip; it allows a proper compaction during pressing and  
103 bestows mechanical strength on unfired tiles (various types of clay materials are used).

- 104 • *Flux* forms a liquid phase at high temperature; it fosters the body densification by viscous flow and  
105 gives rise to a vitreous phase embedding filler (typically various feldspathic materials are used).
- 106 • *Filler* acts as skeleton helping to control the tile shrinkage during drying and firing; it damps  
107 pyroplastic deformations by increasing the body viscosity at high temperature (quintessentially,  
108 quartzous materials are utilized).
- 109 • *Additives* are able to modulate single issues of the technological behavior (e.g., slip rheology,  
110 green density, dry bending strength, firing kinetics, pyroplasticity, color after firing) even if added  
111 in small amounts.

112 Any waste addition will affect the technological behavior in a way that can witness the prevailing role  
113 in the bodies (Segadães, 2006). This is important to correctly address the incorporation strategy,  
114 since every tile type has its own recipe in terms of plastic, flux, and filler ingredients (Fig. 2A).

115 A fundamental requisite for waste recycling is the inertization of residues, once incorporated in the  
116 ceramic body. It means that physical and chemical reactions occurring during the tile production  
117 must result in the immobilization of all the waste components, at least the hazardous ones. This is a  
118 key point, because the conditions to get satisfactory results may be in contrast with either  
119 technological or inertization purposes. For instance, a firing schedule able to achieve the standard  
120 technical requirements (e.g., the prescribed water absorption) could induce a partial mobilization of  
121 a given component of the waste material.

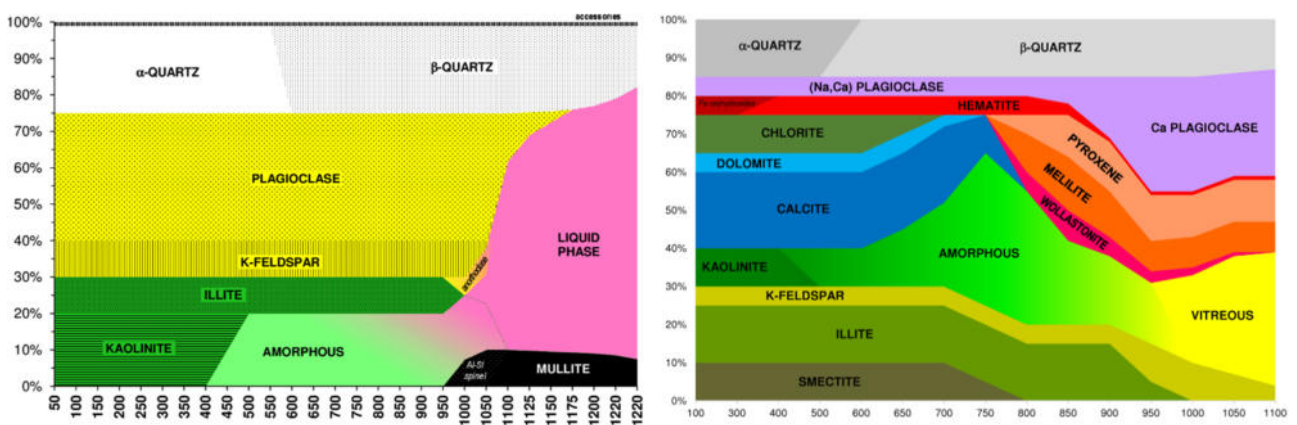


122  
123 Figure 2. Batch design of the various typologies of ceramic tiles (modified after Dondi et al., 2014): light-firing  
124 and red-firing bodies (A). Phase composition of fired bodies: amount of residual phases versus the amorphous-  
125 to-crystalline ratio in phases formed during firing (B).



126 This aspect has been to a large extent neglected in ceramic tile production, under the assumption  
 127 that natural raw materials do not contain hazardous components in concentrations justifying a special  
 128 care. The only exception is about lead and cadmium release from products intended to come in  
 129 contact with foodstuff (ISO 10545-15, 1995). This assumption is no longer valid in case of waste  
 130 materials since they may contain hazardous components in significant concentrations. A successful  
 131 inertization depends on the transformations which the individual components of wastes may (or may  
 132 not) undergo during firing. Such reactions involve the formation of new phases (either crystalline or  
 133 amorphous). As a consequence, the effective incorporation of a given element implies different  
 134 crystal-chemical factors whether the target is a certain silicate structure or a vitreous network. A  
 135 distinctive picture arises for the various technologies (Fig. 2B): recycling in porous and highly porous  
 136 bodies requires a strategy of waste incorporation mainly addressed to crystalline phases. On the  
 137 other hand, recycling in vitrified and highly vitrified bodies must account for the predominant  
 138 development of glassy phases.

139 The reactions leading to the formation of new crystalline and amorphous phases occur at various  
 140 temperatures and define an extremely dynamic picture, governed by kinetic-related issues, such as  
 141 thermal rate and tile geometry (size and thickness) along with the batch mineralogical composition  
 142 (Cultrone et al., 2001; Gualtieri, 2007; Heimann and Maggetti, 2019; Conte et al., 2020). A general  
 143 sketch of phase transformations occurring during fast firing can be drawn, based on literature data,  
 144 for vitrified and porous bodies, respectively (Fig. 3).



145 Figure 3. Scheme of phase transformations occurring during fast firing of ceramic tiles, based on literature  
 146 data: (left) vitrified bodies, like porcelain stoneware; (right) porous bodies, such as red monoporosa-birapida.

147 *2.3. Effect of waste recycling in the manufacturing stages*

148 This section will provide some insight into the main technological issues that may arise with the  
149 addition of waste materials in the various stages of the ceramic tile manufacturing process.

150 *Body formulation:* raw materials must comply a set of requisites, including the physical status at the  
151 delivery, which may determine in many cases a sort of “supply gap” (i.e., all the operations necessary  
152 to turn the physico-chemical features of the waste within the acceptance window for ceramic tile  
153 manufacturing). From this standpoint, the main issues are moisture, grain size, and stability during  
154 storage (in order to avoid phenomena of biodegradation, cementation, leaching, etc). The existence  
155 of a supply gap implies the need for additional steps in the technological chain (the most common  
156 are drying or comminution) hence of deputed machinery and related investment.

157 *Milling* constitutes one of the main bottlenecks for waste recycling, particularly in case of wet  
158 grinding. Any deleterious effect of waste on the rheological behavior of slips must be avoided,  
159 especially thickening of the suspension by viscosity and/or thixotropy boosting. In any case, common  
160 adjustments – like increasing the water or deflocculant amount – imply increasing cost (energy  
161 consumption or additives). Most relevant is the fact that uncontrolled changes of slip viscosity may  
162 result in a different particle size distribution of powders, which in turn can affect the technological  
163 behavior downstream, and particularly the firing kinetics. Unexpectedly, the rheological behavior of  
164 waste-bearing slips has been seldom investigated in detail ([Andreola et al., 1992a, 1992b, 1993,](#)  
165 [2001, 2002, 2004](#)). Critical parameters that should be kept under control for their deleterious effect  
166 are the occurrence of electrolytes (soluble salts), expanding clay minerals (especially smectites),  
167 zeolites, and colloidal fractions.

168 *Compaction* is important as it affects properties of both semi-finished and finished products. Among  
169 others, it substantially determines the extent of firing shrinkage and thus any deviation from the  
170 nominal dimension of tiles (that is a commercial constraint). Such a behavior was considered  
171 occasionally, directly as bulk density or indirectly as modulus of rupture of dry tiles (e.g., [Matteucci](#)  
172 [et al., 2002; Bourtsalas et al., 2015b](#)). Springback was rarely taken into account, even though it can  
173 induce, whether too high, defects and rupture of green compacts (especially thin tiles). Plasticity of  
174 the body was sometimes considered in case of extrusion (e.g., [Menezes et al., 2005; Souza et al.,](#)  
175 [2011](#)). As densification in the green state is promoted essentially by “plastic” components (primarily

176 clay minerals) and contrasted by “non-plastic” ones (fillers and fluxes), a correct ratio between plastic  
177 and non-plastic ingredients must be respected.

178 *Drying* stage is crucial for split tiles, since extrusion is usually carried out with a water content around  
179 20%wt, thus approximately 3-4 times the moisture content of dry pressed tiles. In contrast, green  
180 pressed tiles have a sufficiently low moisture content to stay within the pendulum stage of drying,  
181 where negligible drying shrinkage is expected ([Ford, 1986](#)). Although small, drying shrinkage must  
182 be kept under control and the effect of waste addition monitored. Most important is the dry modulus  
183 of rupture, which must be high enough, because unfired tiles must withstand firstly the stresses  
184 during fast drying cycles, usually peaking around 180°C, and secondly the drop of mechanical  
185 strength due to wetting during glazing. Drying behavior was sometimes taken into account, directly  
186 as drying shrinkage or indirectly as modulus of rupture of dry tiles (e.g., [Raimondo et al., 2007](#);  
187 [Echeverrigaray et al., 2016](#)). Drying is eased by non-plastic ingredients (fluxes and fillers) but  
188 contrasted by plasticity providers, which on the other hand improve bending strength of dry tiles.

189 *Glazing and decoration*: potential interaction of wastes with glazes and decoration is systematically  
190 neglected in the literature. In reality, it is well known in the industrial practice that the occurrence of  
191 some compounds can cause defects to the tile coatings ([Bresciani, 2003](#)). The only issue  
192 considered, for unglazed bodies, is a possible change of color after firing due to the waste addition.  
193 Nonetheless, it must be taken into account that color not only depends on the concentration of  
194 chromophores (typically transition metal ions) supplied by wastes, but it is also affected by the  
195 degree of translucency of the ceramic body. This property is influenced primarily by the amount of  
196 vitreous phase ([Ozturk et al., 2008](#); [Salem et al., 2009](#)).

197 *Firing* effect on the properties of tiles has been extensively considered in the literature, mainly as  
198 shrinkage, water absorption and flexural modulus of rupture. Less frequently, bulk density, closed  
199 porosity and microstructure have been investigated. Firing shrinkage depends on mechanism and  
200 extent of sintering but on the starting porosity as well. It is hard to infer whether any shrinkage  
201 variation induced by waste addition is due to the sintering degree or to an effect of green bulk density,  
202 in absence of starting porosity data. Thus, a critical issue is how to assess the degree of sintering.  
203 Water absorption is commonly determined: although mandatory for standard classification, it is  
204 inconclusive about the actual extent of densification. For vitrified tiles, most important is bulk density,

205 possibly combined with closed porosity. For porous bodies, mechanical strength is a common tool  
206 to evaluate the degree of sintering, since firing reactions progressively strengthen solid bridges  
207 between pores. Nevertheless, it should be appraised by relating the modulus of rupture to the total  
208 porosity (i.e. open and closed pores).

#### 209 *2.4. Design of experiments for waste recycling in ceramic tiles*

210 A correct design of experiments is crucial to get useful results for a technological transfer to the  
211 industrial practice. This task implicates different aspects to be opportunely taken into account.

212 *Technological reliability:* data about the behavior during processing are reliable (and straightly  
213 transferable to industry) if the experimental conditions are as close as possible to the actual industrial  
214 technology. Caution is recommended in data interpretation, especially extending conclusions based  
215 on laboratory results to full-scale processing. Testing bodies in conditions far from industrial practice  
216 – e.g., extrusion instead of pressing or slow firing cycle instead of a fast one – makes the results of  
217 little technological value.

218 *Scale factor:* data reliability depends also on the scale of the experiment: some features of the  
219 industrial manufacture cannot be faithfully reproduced in the laboratory. Classic examples are: firing  
220 atmosphere (combustion fumes in industrial kilns; air in electric furnaces) and green bulk density  
221 (increasing with decreasing die dimensions). Thus, the scale factor is critical for the reproducibility  
222 of results at the industrial level.

223 *Benchmark:* an internal or external reference is of the utmost importance to have a correspondence  
224 with the industrial production and/or market requirements. A benchmark is an effective way to take  
225 care of the scale factor, for instance testing at the laboratory or pilot scale the waste-free batch used  
226 in the industrial production (internal reference). Differences in technological behavior or technical  
227 performance, stemming from waste addition, are usually scaled up according to the benchmark ratio  
228 (lab scale to full scale). The alternative of assessing the laboratory results with reference to standard  
229 specifications is not as straightforward as it may appear, because it is prone to a scale mismatch  
230 that must be estimated anyway.

231 *Recommended amount of recyclable waste:* it is the optimal amount of waste that can be introduced  
232 into a given batch with tolerable changes of technological and environmental issues. This definition  
233 is often disregarded in the literature by weighting in different way various technological outcomes.

234 Conclusions can be too optimistic if positive results are highlighted, while drawbacks tend to be  
235 underestimated, when not neglected. This practice can turn the range of recommended amounts too  
236 extended towards high percentages than the actual chances in industrial production. A common  
237 oversight is claiming the feasibility of waste recycling, even though not adequately supported by  
238 experimental results. For example, the technological assessment, when based uniquely on the firing  
239 behavior, does not ensure an actual feasibility of waste recycling at the industrial level (neglecting  
240 any issue upstream).

### 241 **3. Level of knowledge on waste recycling in tile-making**

242 The literature on waste recycling in ceramic tile-making consists of approximately 300 papers. It is  
243 definitely the tip of the iceberg, because many trials have been carried out directly by tile  
244 manufacturers and suppliers, which results are seldom accessible. Hereafter, three general issues  
245 are overviewed: waste sources, strategies of incorporation, and level of knowledge achieved about  
246 the effect in tile-making.

#### 247 *3.1. Waste sources*

248 The provenance of wastes used by or proposed to the ceramic tile industry is summarized in [Table](#)  
249 [2](#), where the number of papers is also reported. However, it is not always possible to identify, from  
250 the literature, the exact waste classification. For example, soda-lime glass, an apparently well-  
251 defined residue, can be recovered as: i) pre-consumer scrap (a glass bottle) and from at least three  
252 stages of the municipal solid wastes processing: ii) washed glass (secondary raw material for glass-  
253 making), iii) beneficiated glass from contaminated lots, iv) waste-of-waste tailings after beneficiation.  
254 Each waste has its own code, even if it is ultimately soda-lime glass.

255 In any case, the present paper considers recycling under a technological viewpoint (i.e., looking at  
256 waste composition and behavior in ceramic tile manufacturing) and not in relation to waste  
257 classification. Nevertheless, this classification outlines issues that must be taken into account,  
258 because they have very important repercussions (according to the legal framework in every country)  
259 on waste management and the actual chance to convert a given residue into a secondary raw  
260 material ([Schreck and Wagner, 2017](#)).

Table 2. Provenance of wastes used by or proposed to the ceramic tile industry.

**Mining, quarrying and treatment of minerals** (#68) of which: excavation, washing and cleaning of minerals (01 01 02, 01 04 09, ANH; 01 04 10, MNH; #19); tailings from mineral processing (01 03 06, 01 04 12, MNH, #26); ornamental stone cutting and sawing (01 04 13, MNH, #23).

**Preparation of food of animal origin** (02 02 03, ANH, #2): eggshell, cockle shell.

**Production of paper and leather** (#7) of which: rejects from pulping of wastepaper (03 03 07, ANH, #4); grit and lime mud (03 03 09, ANH, #2); sludge from effluent treatment containing chromium (04 01 06, ANH, #1).

**Oil refining** (#8) of which: oily sludge from petroleum refining (05 01 06, AH, #6); sludge from palm oil refining (02 03 01, ANH, #2).

**Inorganic chemical processes** (#10) of which: red mud from bauxite processing (01 03 09, MNH; 01 03 10, MH, #5); Ca-based reaction wastes from TiO<sub>2</sub> production (06 11 01, ANH, #2); waterglass waste (06 03 14, MNH, #1); phosphogypsum (06 09 04, MNH, #1); ferrite production (08 02 02, ANH, #1).

**Thermal processes, coal power stations** (#30) of which: fly ash (10 01 02, ANH, #24) and bottom ash, slag and boiler dust (10 01 01, ANH, #6).

**Thermal processes: biomass combustion plants** (10 01 03, 10 01 15, 10 01 17, ANH, #25) of which: wood, cereal and straw ash (#7); rice and coffee husk ash (#6); sugarcane and coffee bagasse ash (#5); palm kernel shell and sago ash (#3); fishbone and cow bone ash (#3) paper ash (#1).

**Metallurgy, casting and coating of metals** (#37) of which: iron and steel unprocessed slag (10 02 02, ANH, #21) and wastes from slag processing (10 02 01, ANH, #2); copper slag (10 06 01, ANH, #2); zinc slag (10 05 01, ANH, #1); lead slag (10 04 01, AH, #1); foundry slag (10 09 03, ANH, #1); spent foundry sand (10 09 07, MH; 10 09 08, MNH, #7); galvanic processes (11 01 09, MH, #2).

**Manufacture of ceramic products, refractories and glassware** (#40) of which: fired scraps (10 12 08, ANH, #10); green tiles and dust (10 12 01, 10 12 03, ANH, #8); polishing sludge (#6); sludge from wastewater treatment (10 12 13, ANH, #5); glazing sludge and waste frit (10 12 11, 10 11 11, MH, #5); spent lime from kiln scrubbers (10 12 09, MH, #3); glass polishing and cutting (10 11 14, MNH, #2); spent refractories (16 11 06, MHN, #1).

**Construction and demolition wastes** (#12) of which: construction materials containing asbestos (17 06 05, AH, #3); harbour dredging spoil (17 05 05, MH, #2); reservoir sediments (17 05 06, MNH, #2); cement kiln dust (10 13 07, ANH, #2); flat glass (17 02 02, MNH, #2); mixtures of concrete, bricks, tiles and ceramics (17 01 07, ANH, #1).

**Waste management facilities and wastewater treatment** (#37) of which: bottom ash and slag (19 01 12, MNH, 19 01 11, MH, #8) and fly ash (19 01 13, MH, #5) from municipal solid waste incineration; sewage sludge (19 08 05, ANH, #8) and sewage ash (19 01 14, MNH, #7) from treatment of urban wastewater; sludge from treatment of drinking water (19 09 01, ANH, #5); sludge from treatment of industrial wastewater (19 02 06, MNH, #2); vitrified waste (19 04 01, ANH, #2).

**Municipal and industrial solid wastes: separately collected fractions** (#64) of which: container glass (19 12 05, ANH; 20 01 02, MNH, #35); glasses from WEEE (20 01 36, MNH): PC-TV screen (#8); PC-TV cathodic tube (#7); lamps (#5); LCD (#3); other glasses (#6).

262

<sup>1</sup>Codes from the List of Wastes (LoW) annexed to the technical guidance on the classification of waste (2018/C 124/01).

263

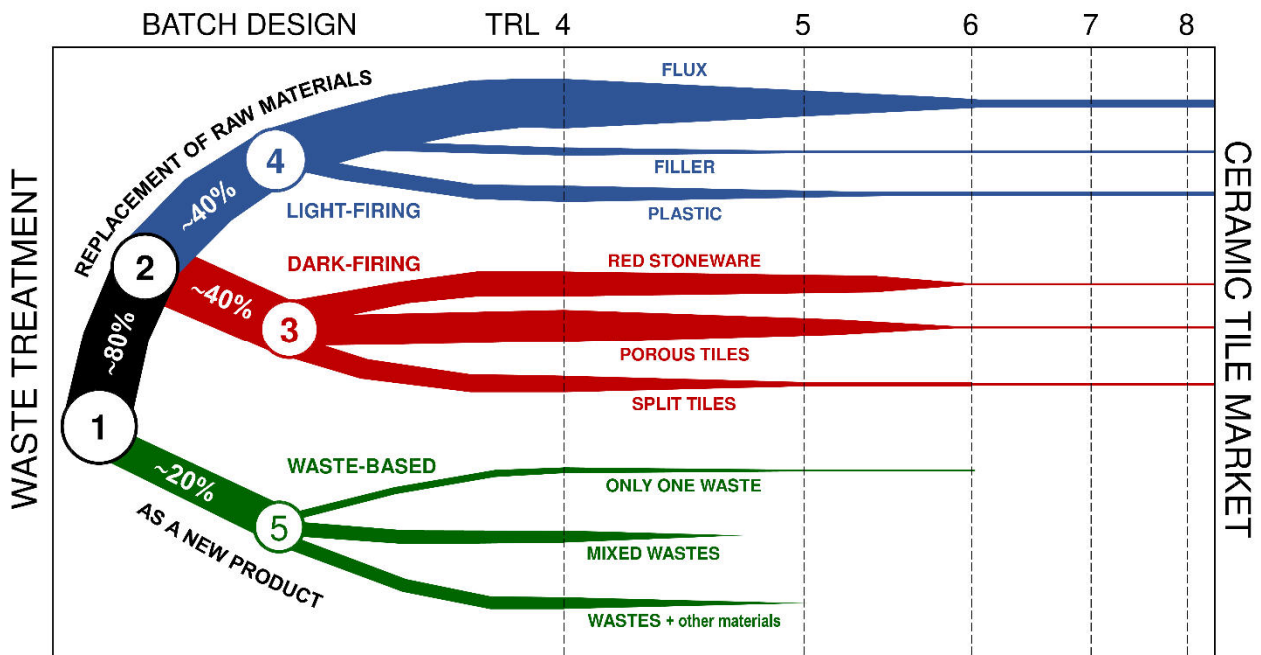
<sup>2</sup>Statement of hazard from the above-mentioned LoW: Absolute hazardous (AH); Absolute non hazardous (ANH); Mirror hazardous (MH); Mirror non hazardous (MNH).

264

265 The main issue recalled here is the fundamental divide between hazardous and non hazardous  
 266 wastes (Blackman, 2001; Sadala et al., 2019). The legal restrictions to recycling (treatment, trade,  
 267 transport, end use) make unappealing for the ceramic tile industry most hazardous wastes. For this  
 268 reason, the attention of industrialists is focused on non hazardous residues only (with few  
 269 exceptions).

270 3.2. Strategies of waste incorporation in ceramic bodies

271 Literature reveals that waste recycling has been addressed to the ceramic tile production through  
 272 different strategies, which clearly stand out from the milestones in batch design (Fig. 4). Most  
 273 research efforts were spent to test wastes in replacement of raw materials conventionally used in  
 274 tile manufacturing. Even so, ~20% of papers dealt with innovative ceramics, based uniquely on waste  
 275 materials (milestone 1). In case of substitution of natural raw materials, batches were designed either  
 276 for dark-firing bodies or light-firing ones, with a similar number of papers (milestone 2). The former  
 277 is an obvious choice when the waste has a high content of transition elements, which are responsible  
 278 for the dark color after firing. For the most part they are clay bodies with minor ingredients playing  
 279 as filler (or flux) in red stoneware, otherwise as filler (or pore-forming agent) in porous tiles. At  
 280 milestone 3, some papers followed processing parameters valid just for extruded split tiles, which  
 281 can be either porous or vitrified.



282  
 283 Figure 4. Strategies followed in the literature for waste recycling in ceramic tiles: from waste treatment to the  
 284 tile market. Milestones in batch design: 1) waste as a new product or as a substitute of natural raw materials.  
 285 2) Light-firing versus dark-firing bodies. 3) Dark-firing bodies for red stoneware, porous or split tiles. 4)  
 286 Technological role in light-firing batches (flux, filler, plastic component). 5) Waste-based products: 100% waste,  
 287 or mixture of different residues, or waste plus other materials (mix different from industrial batches). See the  
 288 text for TRL description.

289 On the other hand, light-colored bodies are obtained by complex batches, where the main  
290 technological role must be guessed at the moment to introduce the waste in replacement of a given  
291 type of raw material. Literature trials saw the wastes mainly substituting a flux and less frequently a  
292 plastic component or a filler (milestone 4). Amongst the waste-based products, three options were  
293 followed (milestone 5): testing the residue as the unique batch ingredient; in a mixture of different  
294 residues (and no other ingredients) or mixed with industrial minerals (in mix completely different from  
295 industrial formulations).

296 The degree of development of the various waste recycling solutions is sketched in [Figure 4](#), in terms  
297 analogous to the Technological Readiness Level (TRL). It is scaled as follows: TRL 4 stands for  
298 waste recycling evaluated at the laboratory scale with processing conditions far from the industrial  
299 production. TRL 5 indicates a technological solution validated at the lab scale in conditions close to  
300 the current tile production. TRL 6 represents a demonstration on a pilot line (industrially relevant  
301 environment). TRL 7 entails a waste-bearing tile prototype successfully obtained in operational  
302 environment (small-scale production). TRL 8 is a waste recycling technology complete, qualified,  
303 and ready for the market.

304 Looking at products consisting only (or mostly) of residues, it clearly emerges that have been  
305 developed essentially at TRL 4. In few cases, a higher TRL was achieved, as for mining residues at  
306 TRL 5 ([Campos et al., 2004](#)), fly ash at TRL 6 ([Luo et al., 2017b, 2018](#)) and glass cullet at TRL 7  
307 ([Skerratt, 2001](#)).

308 In contrast, wastes used in replacement of raw materials were often tested at TRL 5. In some cases,  
309 trials were run up to TRL 8 or even entered in the industrial practice, at least in some ceramic tile  
310 districts. Apparently, this goal was achieved more frequently with light-firing bodies than with dark-  
311 firing ones ([Fig. 4](#)). In reality, literature data mirror more the academic and industrial interest than  
312 the actual technological prospect. It is undoubted that porcelain stoneware is the main target of waste  
313 recycling, accounting for more R&D studies than any other tile type. Successful examples of full-  
314 scale waste recycling in ceramic tile manufacture include: excavation and treatment of industrial  
315 minerals, such as waste granite ([Dino et al., 2012](#)) and residue of sand washing ([Danasino and Di  
316 Primio 1995](#)); glass cullet and glasses from WEEE ([Carbonchi et al., 2003](#); [Andreola et al., 2016](#));  
317 stabilized bottom ash from municipal solid waste incineration ([Schabbach et al., 2012](#)). Nevertheless,



318 the largest amount of residues recycled takes origin from the ceramic factory itself and is  
319 cannibalised in tile-making, encompassing green tiles, dust, sludges from wastewater treatment and  
320 fired scraps (Manfredini et al., 1991; Andreola et al., 2004; García-Ten et al., 2015; Rambaldi et al.,  
321 2018; Tarhan et al, 2017; Amin et al., 2019).

### 322 *3.3. Knowledge of waste effects in tile manufacturing*

323 The level of knowledge on waste addition to ceramic tiles, as it can be deduced from the literature,  
324 is extremely variable. A matrix illustrates at the glance, by a simple color code, the degree of  
325 technological information on waste recycling for single processing steps: partial or full  
326 characterization, its robustness and level at which experiments were carried out (Fig. 5).

327 Looking at this matrix by columns, it appears that information on firing is richer than about all the  
328 other manufacturing stages, despite they are as crucial as sintering to define the product quality  
329 (e.g., milling). The lack of data is quite common about the rheological behavior of slips and powder  
330 behavior during compaction. Unfortunately, these stages frequently represent the bottleneck in the  
331 waste recycling scale up from lab to factory. Apparently unimportant, the need of preliminary  
332 treatments is indeed fundamental to make the waste recycling viable, but information is seldom  
333 exhaustive on this point.

334 The technological know-how is clearly different from one waste source to another. There are cases  
335 where a full characterization (or a partial characterization by multiple sources) is available, such as  
336 ceramic residues or waste glasses, to which generally correspond full scale trials (or at least on a  
337 pilot line). In other circumstances, the firing behavior is well-known, although at the lab scale, but  
338 information on upstream processing is scarce (as for many mining residues, metallurgical slags,  
339 municipal solid wastes and sludges from inorganic chemical processes and oil refining). There are  
340 peculiar situations where, in face of rather numerous data, technological information (not only at the  
341 lab scale) appears to be largely contradictory, as in case of many ashes from thermal processes.  
342 The remaining waste sources suffer from a partial technological characterization, limited to lab  
343 experiments, with few exceptions (e.g., foundry sands and asbestos-containing demolition residues).



344

345 Figure 5. Matrix of technological information available for waste recycling in ceramic tiles. Color codes  
 346 represent: full characterization by multiple sources (4); full characterization by a single source or partial  
 347 characterization by multiple sources (3); partial characterization by a single source and/or contradictory data  
 348 by multiple sources (2); characterization not relevant for ceramic tiles (1); no data (0). Experimentation carried  
 349 out up to: L) laboratory scale; P) pilot line or lab scale approaching industrial conditions; F) full scale or  
 350 industrial plant.

#### 351 **4. Waste composition and recycling strategies in tile-making**

352 This chapter describes the chemical composition and physical state of wastes along with the batch  
353 design in different ceramic tiles. In-deep discussion on the effects of waste addition on the  
354 technological properties of ceramic tiles and the recommended amount of recyclable waste are  
355 reported in the supplementary materials.

##### 356 *4.1. Wastes from mining, quarrying and treatment of minerals*

357 There are many case-studies dealing with recycling in ceramic tiles of mining residues but, despite  
358 the good number of papers, the level of knowledge is essentially incremental and limited to laboratory  
359 scale testing. Waste recycling was mainly addressed to floor and wall tiles (mostly red firing) with a  
360 limited concern to porcelain stoneware bodies. Several papers adopted firing conditions far from  
361 current technologies in tile-making. These residues can be distinguished upon the stage in which  
362 they are produced: (a) during excavation; (b) after washing and cleaning of minerals; (c) as tailings  
363 of beneficiation processes; (d) after cutting and sawing of ornamental stones. Besides these  
364 operations are industrially well established, quarrying and mining residues are characterized by a  
365 strong compositional heterogeneity, which reflects the variety of parent rocks.

366 *Wastes generated during excavation* (mostly discarded blocks and rock dust) have the same  
367 composition of parent rocks and are generally utilized on site for fillings and environmental  
368 remediation ([Table 3](#)). Two exceptions concern ceramic raw materials: granite blocks accumulated  
369 around quarries and marble dust. The best example of successful valorization of waste granite  
370 entails block recovery from quarry dumps and various treatments: grinding, sieving, milling, magnetic  
371 separation, flotation ([Dino et al., 2012](#)). The products so obtained have been commercialized in the  
372 last two decades as flux in bodies for vitrified tiles ([Carbonchi et al., 2003](#)). This was confirmed by  
373 further case-studies on granite ([Ghiani et al., 1997](#); [Hernández-Crespo and Rincón, 2001](#);  
374 [Echeverrigaray et al., 2016](#)) or alkalisyenite mining residues ([Il'ina and Lebedeva, 2010](#)) tested as  
375 flux substitutes in ceramic batches (10-35%). Carbonate-rich residues were proposed as pore-  
376 forming additive in porous tiles in substitution of calcite or dolomite: marble rock dust ([Montero et al.,](#)  
377 [2009a](#)); dolostone ([Amin et al., 2011](#)) and magnesite ([Binal and Ay, 2014](#)).

Table 3. Chemical composition of residues from mining, quarrying and mineral treatments used in (or proposed for) ceramic tile manufacturing.

% wt.	Granite quarrying and beneficiation (6)		Quartzite quarrying, washing and cutting (11)		Ornamental granite sawing & cutting (14)		Shales, kaolin and clay treatment (10)		Ore beneficiation tailings (18)		Borate beneficiation tailings (20)		Marble & limestone quarrying & cutting (8)						
	min	max	min	max	min	max	min	max	min	max	min	max	min	max					
SiO <sub>2</sub>	54.56	74.85	69.30	78.14	53.07	77.92	65.98	44.23	63.88	55.05	6.63	73.20	42.76	0.39	50.48	17.46	0.76	44.50	13.95
TiO <sub>2</sub>	0.11	0.90	0.41	0.56	0.09	0.83	0.40	0.12	2.10	1.11	0.01	11.00	2.07	0.01	0.46	0.14	<0.01	0.21	0.07
B <sub>2</sub> O <sub>3</sub>														3.99	31.80	17.30			
Al <sub>2</sub> O <sub>3</sub>	13.30	18.63	14.59	10.89	11.77	17.49	14.11	16.30	48.66	27.94	2.18	37.19	13.09	0.11	12.58	3.25	0.27	6.00	1.95
Fe <sub>2</sub> O <sub>3</sub>	1.13	7.33	3.49	3.05	0.46	11.66	5.10	0.93	8.42	3.99	0.40	83.04	21.39	0.13	4.24	1.03	<0.01	6.26	1.29
MgO	0.23	1.38	0.63	0.79	0.09	2.62	0.93	0.20	2.28	0.89	0.01	24.57	5.08	0.60	20.84	10.49	0.19	40.37	8.74
CaO	1.02	3.50	2.07	0.61	0.62	7.93	3.84	0.10	2.83	1.10	0.06	22.20	4.98	4.30	52.75	20.25	2.30	57.99	40.75
MnO	0.05	0.20	0.12	0.02	0.03	0.14	0.07				<0.01	0.93	0.17		12.52				
CuO											<0.01	0.50	0.05						
SiO														0.01	1.09	0.17			
Na <sub>2</sub> O	3.00	6.50	3.79	0.25	2.70	4.48	3.38	0.01	1.82	0.43	0.13	8.12	2.06	0.01	16.25	3.30	<0.01	0.74	0.18
K <sub>2</sub> O	3.80	5.62	4.67	2.76	2.58	8.50	4.42	0.24	5.72	2.59	0.07	5.84	2.17	0.01	3.59	0.80	<0.01	0.44	0.13
P <sub>2</sub> O <sub>5</sub>	0.05	0.30	0.18	0.36	0.05	0.75	0.30				0.00	15.10	2.03	0.01	0.04	0.01	<0.01	0.02	0.01
SO <sub>3</sub>				0.07						0.11					0.01	1.10	0.16		
L.o.I.	0.10	1.30	0.55	5.89	0.09	4.44	1.62	5.40	14.50	7.99	0.20	17.73	5.92	11.30	35.16	25.90	15.50	40.00	32.45

Granite quarrying: Ghiani et al., 1997; Hernández-Crespo & Rincón 2002; Ilina & Lebedeva 2010; Dino et al., 2012; Echeverriagaray et al., 2016; Quartzite quarrying and cutting: Menezes et al., 2005; Torres et al., 2007; Correia et al., 2009; Souza & Costa 2015; Biff & Silva 2016; Medeiros et al., 2017; Karhu et al., 2019. Granite sawing & cutting: Hernandez-Crespo et al., 2002; Torres et al., 2004, 2007; Menezes et al., 2005, 2008; Vieira & Monteiro, 2006; Hojamberdiev et al., 2011; Souza et al., 2011; Gadioli et al., 2012; Junkes et al., 2012; Aguiar et al., 2014. Shale and kaolin treatment: Menezes et al., 2005, 2008; Varela et al., 2009; Melnick et al., 2010; Dal Bò & Hoza, 2013; Sanchez-Soto et al., 2018. Ore tailings: Kaya et al., 2004; Messegueur et al., 2009; Wei et al., 2009; Yang et al., 2009; Cavalcante et al., 2012; Das et al., 2012; Chen et al., 2013; Mehriban & Sarraf 2015; Solismaa et al., 2018; Tang et al., 2018; Fontes et al., 2019; Karhu et al., 2019; Pashkevitch & Petrova, 2019. Borate tailings: Olgun et al., 2005; Ediz & Yurdakul 2009; Celik et al., 2014; Celik, 2015; Cetin et al., 2015; Özkan, 2017; Kurama et al., 2007; Çiçek et al., 2018; Korç et al., 2018; Tang et al., 2018; Zanelli et al., 2019; Karadağlı and Çiçek, 2020. Marble quarrying and cutting: Montero et al., 2009a; Amin et al., 2011; Binal & Ay 2014; Jordán et al., 2015; Marvila et al., 2018; Solismaa et al., 2018.

379 *Wastes from washing, cleaning, and de-dusting* operations stem from removing the finest particles  
380 and/or settling the coarsest grains. The resulting wastes have the composition of a given fraction of  
381 parent rocks. In general, the expected role in ceramic batches is either as plasticity-provider (fines)  
382 or filler (sands) or pore-forming (carbonates). Clay materials were recovered from sand washing  
383 ([Danasino and Di Primio 1995](#); [Sokolar and Grygarova, 2013](#); [Sanchez-Soto et al., 2018](#)) and after  
384 kaolin ([Menezes et al., 2005, 2008](#); [Varela et al., 2009](#)), slate ([Campos et al., 2004](#)), oil shale  
385 ([Melnick et al., 2010](#)), serpentinite ([Diaz and Torrecillas, 2007](#)) or gold ([Özkan et al., 2010](#)) mining  
386 operations. Despite the chemical composition is on average comparable to clay raw materials ([Table](#)  
387 [3](#)), as suggested by the mainly kaolinite-illite mineralogical composition, there may be limitations due  
388 to the high content in iron oxide or muscovite. Iron-rich clay wastes were addressed to split tiles  
389 ([Sokolar and Grygarova, 2013](#)) and red stoneware ([Melnick et al., 2010](#)) or, in alternative, as small  
390 addition (<10%) in white-firing bodies ([Özkan et al., 2010](#)). Kaolin waste was tested in both porous  
391 ([Menezes et al., 2008](#)) and vitrified tiles ([Menezes et al., 2005](#); [Varela et al., 2009](#)). Agate rejects  
392 were tested in substitution of quartz sand (up to 65%) in porcelain-like batches ([Correia et al., 2009](#)).

393 *Wastes from mineral beneficiation*, which commonly involves a range of physico-chemical processes  
394 (comminution and sieving, magnetic and electrostatic separation, flotation, acid leaching, and so on),  
395 are frequently referred to as *tailings*. These wastes are usually fine-grained and characterized by the  
396 composition of gangue minerals plus possible contaminants deriving from mineralurgical treatments.  
397 This singles out issues that may hinder the utilization of tailings in tile-making: water content, pH,  
398 hazardous components (from ore and/or processing additives) and overall a composition that can  
399 be far from that of ordinary ceramic raw materials. A distinction is useful between tailings from ore  
400 and borate beneficiation ([Table 3](#)).

401 *Tailings from ore beneficiation* include residues from treatment of iron ore ([Chen et al., 2013](#); [Das et](#)  
402 [al., 2000, 2012](#); [Fontes et al., 2019](#)), Au-Cu-Mo ore ([Mehrabian and Sarrafi, 2015](#); [Solismaa et al.,](#)  
403 [2018](#); [Karhu et al., 2019](#)) and coal ([Wei et al., 2010](#); [Meseguer et al., 2009](#)). In addition, tailings from  
404 industrial minerals, as apatite, bauxite, mica, quartz, feldspar, wollastonite ([Kaya et al., 2004](#); [Yang](#)  
405 [et al., 2009](#); [Solismaa et al., 2018](#); [Karhu et al., 2019](#); [Hossain et al., 2019](#)) and gemstones  
406 ([Cavalcante et al., 2012](#); [Pashkevich and Petrova, 2019](#)) were studied. Beyond the chemical and  
407 mineralogical variability from case to case, common traits are fine particle size, high amount of

408 Fe<sub>2</sub>O<sub>3</sub>, MgO and CaO, apart feldspar (Kaya et al., 2004; Karhu et al., 2019) and coal tailings (Wei et  
409 al., 2010; Meseguer et al., 2009). This circumstance restricts to applications in red-firing bodies,  
410 where tailings were tested as batch ingredient up to 70%.

411 *Tailings from borate processing* are always characterized by an important concentration of B<sub>2</sub>O<sub>3</sub> (4-  
412 32%) that strongly affects their technological behavior (Olgun et al, 2005; Kurama et al., 2007; Ediz  
413 and Yurdakul, 2009; Cetin et al., 2015; Celik, 2015; Özkan, 2017; Tang et al., 2018; Korç et al., 2018;  
414 Çiçek et al., 2018; Karadağlı and Çiçek, 2020; Zanelli et al., 2019). Such residues are featured by  
415 borates (tincalconite, ulexite, colemanite) and a high amount of alkali-earths – due to magnesite,  
416 calcite, plagioclase, chlorite – along with clay minerals and quartz. Sulfur may be present in  
417 concentration not compatible with the gaseous emission threshold from kilns (Table 3). These  
418 tailings were tested, generally below 20%, either in wall tiles (Olgun et al, 2005; Kurama et al., 2007;  
419 Cetin et al., 2015; Celik, 2015; Özkan, 2017; Çiçek et al., 2018) or in porcelain stoneware (Ediz and  
420 Yurdakul, 2009; Korç et al., 2018; Karadağlı and Çiçek, 2020; Zanelli et al., 2019).

421 *Sludges from ornamental stone cutting and sawing* are fine-grained and have a composition close  
422 to that of ornamental stones, but contain processing residues, like scraps of grinding media and  
423 additives (flocculant, etc). This justifies higher percentages in Fe<sub>2</sub>O<sub>3</sub> and CaO, if compared with  
424 granite quarrying residues in Table 3, and raises issues related with too high values of pH and/or  
425 iron and calcium oxides (boosting slip viscosity, dark firing color). The compositional range is rather  
426 wide, comprising granite (Torres et al., 2004, 2007; Menezes et al., 2005; Vieira and Monteiro, 2006;  
427 Souza et al., 2008; Hojamberdiev et al., 2011; Silva et al., 2011; de Almeida et al., 2012; Gadioli et  
428 al., 2012; Aguiar et al., 2014), gneiss (Souza et al., 2010a, 2010b, 2011; Junkes et al., 2011, 2012),  
429 quartzite (Souza and Costa, 2015; Biff and Silva, 2016; Medeiros et al., 2017) on one side and  
430 limestone-marble-doloma on the other side (Montero et al., 2009a; Jordan et al., 2015). Thus, the  
431 expected technological role goes from flux to filler, depending on the feldspars-to-quartz ratio, or  
432 even pore-forming agent in case of carbonate-rich residues. Therefore, various strategies were  
433 followed in batch design: adding the sludge up to 75% in substitution of either a clay in a binary mix  
434 for porous tiles (Menezes et al., 2005; Montero et al., 2009a; Gadioli et al., 2012; Jordan et al., 2015),  
435 or flux or filler in a complex formulation for vitrified bodies (Hojamberdiev et al., 2011; Silva et al.,  
436 2011; Junkes et al., 2011, 2012).

437 *4.2. Wastes from the pulp and paper industry*

438 The residues from the cellulose and paper production that have been investigated as ceramic raw  
439 materials consist in paper mill sludge ([Asquini et al., 2008](#); [Maschio et al., 2009](#); [Furlani et al., 2011](#);  
440 [Sahar et al., 2011](#)), cellulose sludge ([Azevedo et al., 2019](#)), and grits ([Siqueira and Holanda 2018](#)).

441 The mill sludge is affected by a certain variability of the composition, depending on source and type  
442 of paper produced. It cannot be recycled as a sludge and must be treated (de-watering, incineration,  
443 milling). Account must be taken that heavy metals (Ba, Cr, Cu, Pb, Sn, Sr, V) can be found in  
444 significant amount in the ash ([Table 4](#)). This residue was tested at the laboratory scale, alone or  
445 mixed with soda-lime glass and clay (10-60%).

446 Other wastes from the pulp and paper industry consist largely of calcium carbonate ([Table 4](#)). Grits  
447 is a noninert solid waste derived from the liquor causticizing step in cellulose production. It was  
448 tested up to 15 wt% in replacement of calcium carbonate in wall tiles ([Siqueira and Holanda 2018](#)).  
449 Another paper residue was incorporated up to 30% into split tiles ([Azevedo et al., 2019](#)).

450 *4.3. Wastes from petroleum and edible oil refining*

451 Oily sludge, a hazardous material produced during oil extraction, was extensively investigated  
452 ([Pinheiro and Holanda, 2008](#); [Pinheiro et al., 2013](#); [Pinheiro and Holanda, 2013a, 2013b](#)). It is  
453 basically a drilling mud, possibly contaminated by petroleum. Thus, the sludge is based on bentonite,  
454 barite (BaO at 5%) plus additives and fragments of drilled rocks, which all contribute to the high loss  
455 on ignition ([Table 4](#)). Tests at the laboratory scale were conducted adding the sludge to a porcelain  
456 stoneware batch in replacement of kaolin up to 5-10% ([Pinheiro and Holanda, 2008](#); [Pinheiro et al.,](#)  
457 [2013](#); [Pinheiro and Holanda, 2013a, 2013b](#)).

458 Another oily sludge, recovered after processing of palm oil, is expected to be a non hazardous  
459 material with a composition compatible with silicate ceramics ([Table 4](#)). It was introduced up to 20%  
460 in a porcelain stoneware for artware ([Salehi et al., 2015](#)) and tested at the lab scale, even in  
461 combination with a waste glass ([Salehi et al., 2016](#)).

Table 4. Chemical composition of residues from various industries used in (or proposed for) ceramic tile manufacturing.

% wt.	Paper-making sludge (4)		Paper waste	Cellulose grit	Petroleum sludge	Palm oil sludge	Ilmenite reaction wastes		Red mud after bauxite processing (4)		Red mud by zinc metallurgy	Phospho gypsum	Construction & demolition wastes	Cement-asbestos based frit	Refractory rollers	Refractory bricks	Ferrite production sludge	Cement kiln dust
	min	max					mean	min	max	mean								
SiO <sub>2</sub>	23.01	29.10	25.40	8.56	41.73	70.0	18.43	7.60	33.57	15.32	40.06	0.61	60.55	24.86	34.98	50.92	0.80	28.75
TiO <sub>2</sub>	0.42	2.45	1.70	0.35	0.52	0.6	56.43	3.36	5.12	4.57			0.37	0.21	0.30	1.08	0.20	0.21
ZrO <sub>2</sub>	0.80	1.97	1.39	0.01			2.30						0.05		3.19	3.19	95.20	0.19
SnO <sub>2</sub>	0.06	0.41	0.24															
HfO <sub>2</sub>																	1.90	
Al <sub>2</sub> O <sub>3</sub>	15.23	17.40	16.62	8.90	10.93	22.0	5.12	16.63	29.89	22.81	5.15	0.15	10.08	4.07	59.37	30.35	0.53	14.43
Cr <sub>2</sub> O <sub>3</sub>	0.91	1.06	0.99															
Fe <sub>2</sub> O <sub>3</sub>	1.99	6.70	4.46	0.75	7.63	5.0	10.37	7.56	44.34	25.64	15.07	0.08	1.41	2.72	0.69	2.06	0.46	2.08
Y <sub>2</sub> O <sub>3</sub>																		
MgO	2.67	15.99	11.53		5.87	3.5	2.31	0.56	1.57	0.90	1.76	<0.01	0.34	9.52	2.59	5.21	0.02	1.40
CaO	18.48	43.13	26.89	79.00	7.76	3.5	1.11	11.36	17.24	13.88	20.30	30.58	15.02	48.34	0.20	2.44	0.11	44.73
MnO			0.05		0.02						0.35			0.06				
CuO			1.37								0.97							
ZnO				0.05								0.04					0.28	
SrO	0.22	0.28	0.25	0.11	0.29													
BaO			1.80		5.03													
PbO	0.04	1.05	0.71								0.61							
Na <sub>2</sub> O	0.20	4.70	2.70		0.44	0.5		3.17	8.12	4.58	5.00	0.03	0.85	0.26	0.50	0.93	0.06	0.66
K <sub>2</sub> O	0.05	4.48	2.05	1.16	0.95	0.9	1.29	0.07	1.76	0.54	0.16	0.03	4.08	0.29	1.28	1.52	0.02	1.30
P <sub>2</sub> O <sub>5</sub>	1.70	4.91	3.84		0.09						0.33		0.18				0.17	
V <sub>2</sub> O <sub>5</sub>	0.86	1.26	1.06															
SO <sub>3</sub>	0.04	0.70	0.35	1.11			2.60				52.20		0.65			0.52		
Cl																		
L.o.I.			2.01	29.08	18.74			7.29	17.73	11.75	10.87		6.24	9.65				6.37

Paper-making sludge: Asquini et al., 2008; Maschio et al., 2009; Furlani et al., 2011. Red mud from bauxite processing: Pontikas et al., 2007a, 2009; Yang et al., 2009; Wang et al., 2018.



#### 463 4.4. Wastes from inorganic chemical processes

464 In this category are comprised residues from industrial production of alumina, titania, zinc,  
465 phosphoric acid, or sodium silicate. Overall, they consist of “muds” equaled by a fine particle size  
466 distribution and high moisture (semi-solid sludges). However, each residue is featured by chemical  
467 traits, inherited by raw materials, which bring about specific challenges for its recycling (Table 4). All  
468 the investigations were carried out at the laboratory scale.

469 Red mud is the main by-product of bauxite processing to get alumina by the Bayer cycle (Pontikes  
470 et al., 2007a; Yang et al., 2009; Wang et al., 2018). The major component is iron oxide together with  
471 alumina, CaO and silica (Table 4). This fact restricts any application to red-firing products. Another  
472 kind of red mud comes from hydrometallurgical processes of zinc ore (Kummoonin et al., 2014). It  
473 has a silicate basis, rich in Ca and Fe oxides (Table 4) with significant concentrations of heavy  
474 metals, like Zn, Pb, As, Cr, Cu, Ni and V (Hua et al., 2017; Khairul et al., 2019). Again, the high  
475 amount of iron addresses recycling to red-firing bodies or to tiny percentages in white-firing batches.  
476 It was tested (60-70%) in a vitrified tile batch together with steel slag and recycled soda-lime glass.  
477 The sludge after sulfuric dissolution of ilmenite-titania slag to produce titania is particularly rich in  
478 TiO<sub>2</sub>, associated to Si, Fe and Al oxides, among others (Dondi et al., 2010). The main issues are the  
479 fair amount of SO<sub>3</sub> (Table 4) and a certain level of radioactivity (Contreras et al., 2014, 2016). It was  
480 tested in red stoneware (up to 50%).

481 Phosphogypsum is the main waste of the phosphoric acid production (Contreras et al., 2018; Huang  
482 et al., 2020). It is composed of calcium sulfate with all the other components below 1% (Table 4).  
483 This feature is not compatible with the silicate batches used in tile-making, essentially because  
484 calcium sulfate starts decomposing already at 900°C (Coronado et al., 2016) with release of sulfur  
485 oxides in the kiln flue gases. In top of this, a safety issue is represented by the known radioactivity  
486 of phosphogypsum (Huang et al., 2020). It was added up to 10% to a red clay.

487 A sludge stemming from the production of sodium silicate was added (up to 20%) in a batch for floor  
488 tiles (Osman et al., 2008). Adsorbent from solvent cleaning and a residue of ferrite processing were  
489 tested as opacifiers in porcelain stoneware (Mestre et al., 2010).

#### 490 4.5. Wastes from thermal processes

491 Thermal power plants produce energy by burning organic matter of various types: coal, sewage  
492 sludge, municipal solid wastes, and biomasses. Combustion processes give rise to residues that can  
493 be basically distinguished between coarse-grained bottom ash and grate slag (e.g., [Barbieri et al.,](#)  
494 [2002](#); [Bourtsalas et al., 2015a](#)) and fly ash that generally has a very fine particle size (e.g., [Dana et](#)  
495 [al., 2005](#); [Wang et al., 2017](#)). The main chemical traits are inherited from coal or other fuels. The  
496 knowledge about the ash effect in ceramic tile bodies is essentially limited to the laboratory scale,  
497 with few examples at a pilot plant (e.g., [Mishulovich and Evanko, 2003](#); [Ji et al., 2016](#); [Luo et al.,](#)  
498 [2017a](#)).

499 *Coal ashes* have been extensively investigated, pursuing various strategies. Despite the remarkable  
500 variability depending on the coal source (Table 5), their chemical composition presents common  
501 features in silica, alumina, CaO and Fe<sub>2</sub>O<sub>3</sub>, and relative scarcity of alkalis, making coal ashes  
502 compatible with ceramic batches, particularly those for porous bodies. A matter of concern is the  
503 amount of some heavy metals (As, Cr, Cu, Ni, Zn) particularly in fly ash ([Pandey et al., 2011](#); [Tsiridis](#)  
504 [et al., 2012](#)). Since the iron content is rather high, the coloration of the ceramic body is generally  
505 dark. Therefore, the target was mainly constituted by bodies for red stoneware, together with porous  
506 red tiles or simple red clay:ash mixes for split tiles. Attempts to recycle coal ash in porcelain  
507 stoneware are rare.

508 *Coal bottom ash* was investigated in substitution of kaolin, pottery stone, feldspar and quartz (up to  
509 50%) in light-firing stoneware ([Prasartsee et al., 2018](#)); in ash:clay (up to 80%) and ash:clay:sand  
510 mixes (up to 40%) for red stoneware ([Namkane et al., 2016, 2017](#)); added (up to 30%) to a porous  
511 red body ([Skoronski et al., 2015](#)) even together with a borate tailing ([Bayca et al., 2008](#)).

512 *Coal fly ash* was tested in replacement of feldspar and quartz (up to 20%) in porcelain stoneware  
513 ([Dana et al., 2005](#); [Kockal, 2011](#)); in substitution (up to 40%) of clay ([Rajamannan et al., 2013](#); [Kim](#)  
514 [et al., 2017](#)) or up to 10% instead of feldspar ([Olgun et al., 2005](#)) in porous tiles; as clay-ash mixes  
515 (up to 90%) in stoneware-like or split tiles ([Mishulovich and Evanko, 2003](#); [Zimmer and Bergmann,](#)  
516 [2007](#); [Yürüyen and Toplan, 2009](#); [Sokolar and Smetanová, 2010](#); [Sokolar and Vodová, 2011](#); [Trnik](#)  
517 [et al., 2013](#); [Michalíková et al., 2014](#); [Ji et al., 2016](#); [Wang et al., 2017](#)).

Table 5. Chemical composition of ashes from thermal processes (coal and municipal solid wastes) used in (or proposed for) ceramic tile manufacturing.

% wt.	Coal fly ash (24)			Coal bottom ash (7)			Municipal solid waste incinerator bottom ash (6)			Municipal solid waste incinerator fly ash (4)			Sewage sludge ash (5)		
	min	max	mean	min	max	mean	min	max	mean	min	max	mean	min	max	mean
SiO <sub>2</sub>	27.90	62.12	<b>50.98</b>	28.60	65.48	<b>42.26</b>	30.31	53.60	<b>38.27</b>	10.53	36.30	<b>20.11</b>	25.40	83.33	<b>51.21</b>
TiO <sub>2</sub>	0.01	2.92	<b>1.01</b>	0.27	1.03	<b>0.50</b>	0.24	1.54	<b>1.13</b>	0.01	3.40	<b>1.80</b>	0.09	0.29	<b>0.19</b>
B <sub>2</sub> O <sub>3</sub>							0.30	0.56	<b>0.43</b>				4.87	40.25	<b>17.71</b>
Al <sub>2</sub> O <sub>3</sub>	9.93	49.88	<b>24.47</b>	12.30	21.86	<b>17.16</b>	9.95	13.46	<b>12.36</b>	4.35	14.00	<b>7.64</b>	0.01	0.22	<b>0.08</b>
Cr <sub>2</sub> O <sub>3</sub>	0.09	0.11	<b>0.10</b>										4.13	31.83	<b>14.44</b>
Fe <sub>2</sub> O <sub>3</sub>	1.96	12.64	<b>6.38</b>	3.22	18.20	<b>7.92</b>	4.00	13.05	<b>7.23</b>	0.98	18.40	<b>6.00</b>			
MgO	0.24	7.52	<b>2.32</b>	0.46	4.16	<b>2.03</b>	2.64	4.67	<b>3.41</b>	0.01	3.40	<b>1.98</b>	1.15	5.46	<b>2.63</b>
CaO	0.53	43.71	<b>9.30</b>	2.81	38.08	<b>19.94</b>	2.30	35.58	<b>23.21</b>	0.01	62.00	<b>28.38</b>	1.41	21.05	<b>6.60</b>
MnO	0.11	2.80	<b>1.02</b>				0.11	0.31	<b>0.18</b>						
CuO							0.11	0.68	<b>0.35</b>						
ZnO							0.26	0.95	<b>0.53</b>				0.28	1.32	<b>0.80</b>
PbO							0.31	0.36	<b>0.33</b>						
Na <sub>2</sub> O	0.10	3.42	<b>0.77</b>	0.21	1.22	<b>0.74</b>	1.94	4.53	<b>3.09</b>	1.10	4.60	<b>2.57</b>	0.48	8.69	<b>3.46</b>
K <sub>2</sub> O	0.08	4.24	<b>1.40</b>	0.96	3.11	<b>1.96</b>	0.94	1.68	<b>1.27</b>	0.21	3.70	<b>2.17</b>	0.78	3.30	<b>1.62</b>
P <sub>2</sub> O <sub>5</sub>	0.07	0.26	<b>0.16</b>				1.26	2.18	<b>1.80</b>				14.20	17.75	<b>15.98</b>
SO <sub>3</sub>	0.10	4.00	<b>1.84</b>	1.09	3.21	<b>2.15</b>	1.01	3.05	<b>1.93</b>	2.20	5.95	<b>4.08</b>			
Cl							0.48	2.27	<b>1.38</b>	0.85	15.26	<b>8.05</b>			
L.o.I.	0.22	11.96	<b>3.64</b>	0.86	12.60	<b>6.55</b>	5.58	22.20	<b>11.80</b>						<b>8.36</b>

Coal fly ash: Olgun et al., 2005; Chandra et al., 2005; Dana et al., 2005; Sokolar, 2007; Zimmer & Bergmann, 2007; Yürüyen & Toplan, 2009; Sokolar & Smetanova, 2010; Kockal, 2011; Sokolar & Vodová, 2011; Trník et al., 2013; Dal Bò & Hotza, 2013; Das et al., 2013; Michalíková et al., 2014; Ji et al., 2016; Kim et al., 2017; Wang et al., 2017; Luo et al., 2017a, 2017b, 2018. Coal bottom ash: Bayca et al., 2008; Dal Bò & Hotza et al., 2013; Michalíková et al., 2014; Skoronski et al., 2015; Namkane et al., 2016, 2017. MSWI bottom ash: Barbieri et al., 2002; Rambaldi et al., 2010; Schabbach et al., 2012; Bourtsalas et al., 2015a. MSWI fly ash: Hernandez-Crespo & Rincon, 2000; Haijing et al., 2007; Jordan et al., 2015; Bourtsalas et al., 2015b. Sewage sludge ash: Lin et al., 2006, 2008, 2016; Merino et al., 2007; Chen and Lin, 2009.

519 In addition, special batches were experimented, based on fly ash only (Luo et al., 2017a, 2018) or  
520 admixed with other residues (Sokolar, 2007; Tan and Wang, 2011) or talc (Chandra et al., 2005).

521 *Sewage sludge ash* is basically a silico-aluminous material, on average rich in  $P_2O_5$  and  $Fe_2O_3$ , with  
522 fair amounts of alkalis and alkali-earths (Table 5). It can have small concentrations of heavy metals  
523 (Cr, Cu, Mn, Zn). In any case, these chemical features are strongly variable according to the sources.  
524 Sewage ash was experimented alone (Lin et al., 2006); added with nanosilica (Chen and Lin, 2009;  
525 Lin et al., 2016); or admixed in high percentage (minimum 25%) to clay materials (Lin et al., 2008)  
526 and further wastes, like soda-lime glass (Merino et al., 2007) or steel slag (Favoni et al., 2005). No  
527 investigation was carried out in industrial tile-making conditions. Preliminary treatments consisted  
528 essentially in drying and grinding the agglomerated ash.

529 *Ashes from municipal solid waste incinerator (MSWI)* are special wastes to be managed in form of  
530 fly ash or bottom/grate ash. Fly ash was utilized (up to 30%) as clay substitute in porous tile (Haiying  
531 et al., 2007; Jordán et al., 2015) or porcelain stoneware (Hernández-Crespo and Rincón, 2001;  
532 Hernández-Crespo et al., 2002). Bottom ash was added up to 20% directly to a porcelain stoneware  
533 batch (Andreola et al., 2001, 2002). In alternative, previously vitrified bottom and grate ashes were  
534 tested in porcelain stoneware, as a replacement (up to 10%) of feldspar (Rambaldi et al., 2010) or  
535 added up to 10% to the body (Barbieri et al., 2002) or to a clay (Schabbach et al., 2012). The same  
536 vitrified bottom ash was also experimented (5%) in a porous tile body (Rambaldi et al., 2010). Finally,  
537 MSWI bottom ash was investigated mixed with up to 30% soda-lime glass (Bourtsalas et al., 2015a)  
538 or added to the sludge stemming from porcelain stoneware tile polishing in the 10:90 ratio, with an  
539 extra 10% to 20% silica addition, to produce lightweight vitrified ceramic tiles (Tan and Wang, 2011).

540 MSWI ashes have a complex composition, intrinsically more variable than other combustion  
541 processes (Table 5). Although the high contents of alkalis and alkali-earths, particularly CaO, can  
542 be exploited to promote sintering, especially in porcelain stoneware tiles, MSWI ashes exhibit  
543 chemical features that heavily constrain their utilization as ceramic raw materials. Apart from the  
544 abnormally low values of silica and alumina, the concentration in iron oxide is a strong limit to the  
545 use in light-firing bodies. More important is to keep under control the modest concentration of heavy  
546 metals (Ba, Cd, Cr, Cu, Hg, Mn, Pb, Sn, Zn) and, above all, the remarkable quantities of chlorine  
547 and sulfur (Zacco et al., 2014; Zhou et al., 2015). These components can turn intolerable in relation

548 to the release by leaching and the gaseous emissions during firing, respectively. MSWI bottom ashes  
549 are typically finer than 4 mm and can be directly mixed with clay and non-plastic ingredients.  
550 Otherwise, a preliminary treatment (e.g., milling, calcination) is needed (Bourtsalas et al., 2015a).  
551 *Combustion of biomasses* produces different ashes, as sources are extremely varied: rice husk  
552 (Dana and Das, 2002; Andreola et al., 2013; Guzmán et al., 2015, 2016; Abeid and Park, 2018),  
553 coffee husk (Acchar et al., 2013a, 2016; Acchar and Dultra, 2015; Carvalho et al., 2017), sugar cane  
554 bagasse (Sivakumar et al., 2014; Schettino and Holanda, 2015; Faria and Holanda, 2016; Paranhos  
555 et al., 2017), palm oil and sago (Noh et al., 2014; Jamo et al., 2015; Aripin et al., 2010), cereal straw  
556 and stalk (Guzman et al., 2013; Amutha et al., 2014), wood and forestry wastes (Olokode et al.,  
557 2013; Novais et al., 2015), fish bone (Naga et al., 2014; Awaad et al., 2015) and leather waste  
558 (Fernandes and Ferreira, 2007). In addition, coffee grounds were investigated (Manni et al., 2019).  
559 Burning biomasses gives rise to ashes with a considerably variable composition, primarily depending  
560 on the source. In general, plant-based ashes are siliceous materials, poor in alumina and rich in  
561  $P_2O_5$  with respect to ceramic bodies (Table 6). The content in alkalis and alkali-earths is rather high,  
562 especially some peaks over 20% CaO and over 10%  $K_2O$ . An exception is fish bone ash, being  
563 essentially a calcium phosphate waste. Some concern should be addressed to the amount of Cl and  
564 S (Niu et al, 2013) as well as to the fate of heavy metals, like Co, Cr, Ni and Zn (Grammelis et al.,  
565 2006). Among the undesired components, iron oxide is sometimes in significantly high amount for  
566 light-firing bodies. Also, sulfur and chlorine can be present in quantities that require careful evaluation  
567 of the risk of polluting emissions during firing. Moreover, biomass ashes may be particularly rich in  
568 organic residues, to the point to need a calcination before recycling (Noh et al., 2014). Recycling of  
569 biomass ash was tried above all in porcelain stoneware bodies, where the waste was introduced in  
570 substitution of feldspar up to 60% (Guzman et al., 2013, 2015), quartz up to 20% (Amutha et al.,  
571 2014; Schettino and Holanda, 2015; Abeid and Park, 2018), clay up to 40% (Olokode et al., 2013;  
572 Abeid and Park, 2018), phyllite up to 10% (Paranhos et al., 2017) or up to 25% as addition to the  
573 whole body (Fernandes and Ferreira, 2007; Sivakumar et al., 2014; Novais et al., 2015).  
574

Table 6. Chemical composition of ashes from thermal processes (biomasses) used in (or proposed for) ceramic tile manufacturing.

% wt.	Sugar cane bagasse ash (4)			Sago ash Aripin et al., 2007	Palm oil ash Noh et al., 2014	Coffee husk ash Carvalho et al., 2017	Rice husk ash Dana & Das, 2002	Rice straw ash Guzman et al., 2015	Wood ash Olokode et al., 2013	Fish bone ash Naga et al., 2014	
	<i>min</i>	<i>max</i>	<b>mean</b>								
SiO <sub>2</sub>	49.19	69.81	62.25	68.03	66.91	15.20	90.23	79.62	67.73	42.85	0.77
TiO <sub>2</sub>	0.38	1.46	0.80	0.03		0.29	0.13				0.04
Al <sub>2</sub> O <sub>3</sub>	5.92	7.97	6.88	6.93	6.44	4.90	1.07	0.27	0.16		0.61
Cr <sub>2</sub> O <sub>3</sub>				0.04							
Fe <sub>2</sub> O <sub>3</sub>	0.92	8.73	5.34	0.76	5.72	1.79	0.27	0.26	0.36	6.13	0.06
MgO	1.17	3.91	2.50	2.39	3.13	14.50	0.18	0.89	2.46	0.65	0.88
CaO	2.63	10.02	6.76	14.88	5.56	38.60	0.39	2.80	1.64	27.93	59.26
MnO	0.05	0.20	0.12	0.65		0.16	0.00	0.71	0.48	2.40	
CuO								0.01	0.01		
ZnO								0.01	0.06	0.44	0.02
SrO						0.27		0.01			0.17
BaO									0.06		
Na <sub>2</sub> O	0.10	0.71	0.41	1.35	0.19		0.00	0.35	0.42	0.67	0.74
K <sub>2</sub> O	2.65	12.36	7.68	1.35	5.20	12.90	1.36	10.53	15.71	4.54	0.11
P <sub>2</sub> O <sub>5</sub>	0.98	2.23	1.41	1.66	3.72	10.80	3.83	1.61	3.51		36.84
SO <sub>3</sub>	0.42	1.73	1.08	2.02	0.33	0.31		1.74	1.09		0.30
Cl						0.14		0.31	1.68		0.08
L.o.I.	3.69	9.78	7.72	2.30	2.30	9.82	3.83	0.59	3.09	13.73	

Sugarcane bagasse ash: Sivakumar et al., 2014; Schettino and Holanda, 2015; Faria & Holanda, 2016; Paranhos et al., 2017.

576 Otherwise, these ashes were added (up to 50%) to clay-based stoneware ([Aripin et al., 2010](#); [Acchar](#)  
577 [et al., 2013a](#); [Naga et al., 2014](#); [Awaad et al., 2015](#)); up to 25% in replacement of quartz in porcelain  
578 batches ([Noh et al., 2014](#); [Jamo et al., 2015](#)); up to 5% instead of clay in wall tiles ([Faria and Holanda,](#)  
579 [2016](#)); as the only ingredient of glass-ceramic tiles ([Andreola et al., 2013](#)).

#### 580 *4.6. Wastes from metallurgical, steel casting and metal coating industries*

581 This category includes mainly slags which physical and chemical properties depend on the metal  
582 being produced and on the solidification process used, including the type of furnace and in particular  
583 the raw materials. These by-products derive from ferrous metallurgy – blast furnace slags (BFS),  
584 electric arc furnace slags (EAFS), basic oxygen furnace slags (BOFS) – and non-ferrous metallurgy,  
585 like copper, lead or zinc slags. In addition, wastes produced from metal casting (spent foundry sand)  
586 and metal coating (galvanic sludge) have been considered. Apparently, metallurgical wastes were  
587 tested without any specific care to pre-treatment, except in the few cases where the preliminary  
588 grinding of slags ([Furlani et al., 2012, 2013](#); [Lim et al., 2016](#)) or a previous calcination of the sludge  
589 ([Reinosa et al., 2010](#)) were carried out.

590 Slags were used either in replacement of clay in simple binary mixes or in substitution of feldspar as  
591 a fluxing agent in porcelain stoneware tiles. Some experiments were addressed to novel bodies,  
592 mostly made up of waste or mixing raw materials under a rationale different from current industrial  
593 tile production. In the literature examined, the industrial tile-making conditions were seldom followed,  
594 and all tests were run at the laboratory scale. Only in few cases a comparison with the technical  
595 performance of commercial tiles was performed.

596 *Blast furnace slags* are the most common by-product of pig iron production, discharged from blast  
597 furnaces in the primary melting process of iron ore. BFS essentially consist of various silicates  
598 (melilite as the major component) and result to be rich in CaO and MgO but poor in silica with respect  
599 to ceramic tiles. Those tested in ceramic batches are usually poor in iron but may be rich in  
600 manganese ([Table 7](#)). BFS were experimented under different strategies: in porcelain stoneware  
601 (12-30%) as a substitute of feldspar ([Dana and Das, 2003](#)) even mixed with BOFS ([Pal and Das,](#)  
602 [2013](#); [Siddiqui et al., 2014](#); [Zhao et al., 2015](#); [Pal et al., 2016](#)) or iron ore tailings and/or coal fly ash  
603 ([Das et al., 1996](#); [Dana et al., 2005](#)).

Table 7. Chemical composition of residues from metallurgical, casting and metal coating processes used in (or proposed for) ceramic tile manufacturing.

% wt.	Blast furnace slag, BFS (8)			Basic oxygen furnace slag, BOFS (4)			Electric arc furnace slag, EAFS (8)			Non ferrous slag		Spent foundry sand, quartz-zircon (4)			Spent foundry sand, olivine		Galvanic sludge
	min	max	mean	min	max	mean	min	max	mean	Abdrakhimov et al., 1990	Naga & El-Maghraby, 2002	min	max	mean	Furlani et al., 2012	Furlani et al., 2013	
SiO <sub>2</sub>	31.54	41.20	34.69	7.95	14.45	9.77	11.64	28.82	19.76	37.28	33.45	51.02	69.03	63.83	55.10	41.35	23.75
TiO <sub>2</sub>	0.58	1.20	0.98	0.75	0.77	0.76	0.18	0.53	0.34		0.67	0.39	0.50	0.45	0.16		
ZrO <sub>2</sub>												11.61	11.75	11.68			
Al <sub>2</sub> O <sub>3</sub>	8.86	20.47	15.92	1.13	2.13	1.61	4.59	14.42	8.81	9.38	7.69	11.95	18.00	13.60	1.89	0.96	0.91
Cr <sub>2</sub> O <sub>3</sub>		0.12					0.25	0.92			1.04				1.40		22.68
Fe <sub>2</sub> O <sub>3</sub>	0.25	8.98	1.78	19.20	29.84	26.75	6.74	42.40	27.78	14.70	16.62	2.29	3.47	2.82	4.76	6.60	1.09
MgO	2.45	9.88	6.74	5.83	9.78	8.76	2.60	12.78	4.83	4.02	7.50	2.31	3.86	3.42	8.85	45.65	3.78
CaO	32.72	38.99	35.50	30.04	44.82	40.56	22.80	42.71	32.21	28.76	19.93	0.90	2.07	1.49	5.25	1.08	15.12
MnO	2.09	6.24	4.17				1.72	4.46	3.38		0.28			0.09			0.11
NiO															1.15	0.27	14.04
CuO											3.67						7.45
ZnO											5.23						5.08
BaO																	1.46
PbO																	1.30
Na <sub>2</sub> O	0.18	1.84	0.77	0.04	0.32	0.19	0.05	0.63	0.24	1.33	0.45	1.93	5.20	3.57	0.41		0.22
K <sub>2</sub> O	0.08	1.29	0.79	0.04	0.21	0.13	0.02	0.82	0.18		0.44	0.47	1.60	1.04	0.74		
P <sub>2</sub> O <sub>5</sub>															0.25		
SO <sub>3</sub>	0.80	2.38								2.42							
L.o.I.	0.23	1.85	0.73		6.14		0.01	3.91	1.13	1.45			25.61		18.00	3.10	

BFS: Das et al., 1996; Dana and Das, 2002; Dana et al., 2005; Mostafa et al., 2010; Siddiqui et al., 2010; Siddiqui et al., 2014; Zhao et al., 2014; Pal et al., 2016. BOFS: Dana and Das, 2002; Sarkar et al., 2010; Hasmaliza et al., 2014; Zhao et al., 2015; Lim et al., 2016; Karayannis et al., 2017; Teo et al., 2019a. Spent foundry sand: Quaranta et al., 2010; Luo et al., 2014; Coronado et al., 2015; Lin et al., 2017.



605 Used also in replacement of limestone and kaolin (6-33%) in wall tiles (Ozturk and Gultekin, 2015)  
606 and admixed from 50 to 100% with kaolin (Mostafa et al., 2010).

607 *Basic oxygen furnace slags* from steelmaking are characterized by a high content of CaO, Fe<sub>2</sub>O<sub>3</sub>  
608 and MgO, as well as particularly low values of silica and alumina with reference to ceramic batches  
609 (Table 7). The most common crystal phases are merwinite, calcium ferrite, and iron oxides. These  
610 features should restrict the target to dark firing bodies rich in alkali-earths, like some porous tiles.  
611 Nevertheless, BOFS were proposed as fluxing agent in porcelain stoneware and triaxial porcelain  
612 batches (6-40%) in replacement of feldspar (Das et al., 2013; Zhao et al., 2014) and even mixed  
613 with BFS (Pal and Das, 2013; Siddiqui et al., 2014; Zhao et al., 2015; Pal et al., 2016) or coal fly ash  
614 (Das et al., 2013).

615 *Electric arc furnace slags* are dense and tough aggregates, which typically contain high amounts of  
616 FeO, CaO, and MgO, associated with rather low percentages of SiO<sub>2</sub> and Al<sub>2</sub>O<sub>3</sub>, with respect to the  
617 raw materials they are expected to replace (Table 7). In addition, small amounts of MnO and Cr<sub>2</sub>O<sub>3</sub>  
618 can be present, along with traces of Ba, V, Pb, and Mo, that may constitute a matter of concern for  
619 recycling (Quijorna et al., 2014; Mombelli et al., 2016). These features, combined with high Fe  
620 content that can be both in metallic and oxide form, require a preliminary treatment to crush the slag  
621 and remove metallic iron. EAFS have been mainly suggested as fluxing agent in slag-based  
622 stoneware batches (10-60%) either substituting feldspar (Dana and Das, 2002; Hasmaliza et al.,  
623 2014; Teo et al., 2014; Lim et al., 2016) or clay (Teo et al., 2019a, 2019b). At variance, EAFS were  
624 tested (30-40%) in a red stoneware batch (Sarkar et al., 2010) or added to red clays (3-40%) for split  
625 tiles (Karayannis et al., 2017).

626 *Non ferrous slags* produced by metallurgy of copper or zinc exhibit chemical characteristics that  
627 somehow fall within the range defined by steelmaking ones. Along with the high concentration of Ca,  
628 Fe, Mg, and the rather low amounts of Si and Al, their distinctive trait is the occurrence of significant  
629 amounts of metals inherited from ores, e.g. Cu or Zn (Table 7). These slags were studied as raw  
630 materials for ceramic tile bodies (Abdrakhimov et al., 1990; Naga and El-Maghraby, 2002) or glazes  
631 (Ozturk et al., 2019) in replacement of clay (10-20%) or dolomite (up to 30%).

632 *Foundry sands* are utilized together with binders (e.g., bentonite) to create molds or cores,  
633 particularly for casting iron-based alloys. High quality quartz, zircon or olivine sands are generally

634 used, depending on the metal under casting. Therefore, foundry sands can be either rich in silica  
635 and zirconium oxide or magnesium oxide and iron oxide, with alumina and other components  
636 provided by bentonite. In addition, olivine contains moderate amounts of nickel, and chromium ([Table](#)  
637 [7](#)). In any case, spent sands are not always recovered separating each type, so that composition  
638 may significantly vary according to the mix. Moreover, spent sands usually retain debris of cast  
639 metals and their impurities, such as Pb, As, Cr, Mo, Ni, Cu, Hg ([Alonso-Santurde et al., 2010](#);  
640 [Siddique et al., 2010](#)). Quartz sand was essentially experimented in clay:waste binary mixes, up to  
641 50% in red clay ([Quaranta et al., 2010](#); [Coronado et al., 2015](#); [Lin et al., 2018](#)) or up to 60% in ball  
642 clay ([Luo et al., 2014](#)). In addition, it was an ingredient of multiwaste bodies, including glass cullet,  
643 metallurgical slag and/or sewage sludge ash ([Coronado et al., 2015](#); [Lin et al., 2017](#)). Similarly,  
644 olivine sands were admixed (20-60%) with different red clays ([Furlani et al., 2012, 2013](#)).

645 *Galvanic sludges* stem from cleaning of wastewater resulting after chemical treatment of metallic  
646 surfaces. Their chemical composition is reknown for a high concentration of hazardous elements,  
647 such as Cr, Zn, Ni, Cu, and Pb ([Karlovic et al., 2008](#); [Ozdemir and Piskin, 2012](#)). Sludges tested in  
648 tile-making are characterized by relatively high amount of lime and scarce presence of silica with  
649 respect to ceramic tile batches ([Table 7](#)). Moreover, they show the presence of crystalline phases,  
650 such as quartz, calcite, and gypsum. This implies a certain amount of gas-forming species, so the  
651 sludge was used only after pre-calcination, which led to the formation of spinel  $\text{NiCr}_2\text{O}_4$  and diopside.  
652 Ash was added up to 20% to stoneware tiles ([Ferreira et al., 1999](#); [Silva et al., 2010](#)) or glass cullet  
653 ([Reinosa et al., 2010](#)).

#### 654 *4.7. Wastes from the production of ceramics, glasses, and refractories*

655 The ceramic tile factories have since long time developed a range of technological solutions to  
656 cannibalistically recycle almost all wastes deriving from the different phases of the production  
657 process ([Timellini et al., 1983](#)). Nowadays, most of the residues produced by the ceramic tile industry  
658 can be reused, as demonstrated by the pluriannual experience of waste management in the  
659 Sassuolo district in Italy ([Busani et al., 1995](#); [Palmonari and Timellini, 2000](#); [Resca et al., 2015](#)). In  
660 the industrial practice, waste recycling is managed empirically, relying more on trial-and-error than  
661 on a design of experiment, and results are just occasionally divulged. In any case, this cannibalistic  
662 loop is made possible by the quite constant composition of wastes that is fully compatible with that

663 of the desired product. The main ceramic residues stem from: purification of waste waters, including  
664 glazing sludges; unfired scraps and dust; scraps of fired products, including waste frit; tile polishing;  
665 spent lime of kiln scrubbers.

666 *Ceramic sludges* come from the treatment of wastewaters, which are collected by washing and  
667 cleaning operations, glaze preparation, condensation in the various driers, and decanted in ponds  
668 (Zannini, 2020). The sludge obtained from water purification can be added directly in the mill, even  
669 if a direct use of wastewaters, without settling, is possible. In general, a sludge content up to 5% was  
670 retained compatible with the industrial requirements (Manfredini et al., 1991; Andreola et al., 2004).  
671 In alternative, the sludge, once previously dried, was tested in substitution (up to 50%) of the batch  
672 for porous tiles (Amin et al., 2019).

673 *Ceramic green scraps* include wastes produced before the firing process (broken tiles, slip and  
674 powder sieving rejects, dust) which basically maintain the chemical and mineralogical composition  
675 of the tile batch (Table 8A). These residues exhibit different physical status and particle size  
676 distribution: broken tiles are made up of irregular scraps or lumps; slip sieving after milling provides  
677 a mud; powder sieving after spray-drying gives clumpy rejects. Dust from spray-drying air purification  
678 is produced from the cyclone that follows the spray drier and is characterized by a smaller particle  
679 size with respect to standard spray-dried powders. Although we are dealing with a widespread  
680 industrial practice, more than for other kinds of waste, scientific papers are scarce (El-Fadaly et al.,  
681 2010; García-Ten et al., 2015; Rambaldi et al., 2018; Valença and Ferraço, 2018; Amin et al., 2019).

682 *Scraps of fired ceramics* represent a composite group in terms of both ceramic material (wall and  
683 floor tiles, sanitaryware, refractories, waste frit) and process generating the waste. They are  
684 originated during tile finishing (cutting, grinding, and squaring) and product sorting (tiles broken or  
685 discarded for intolerable flaws) or by stock management (unsold lots). As a consequence, their  
686 compositions faithfully reflect that of the recycled ceramic material, apart residues of abrasive tools  
687 and glaze coatings, as summarized for porcelain stoneware, stoneware and various sanitaryware  
688 bodies (Table 8B). Fired scraps must be crushed before being reused, in order to match the usual  
689 grain size of raw materials they can substitute: filler and/or flux (Karamanov et al., 2006; Tarhan et  
690 al, 2017; Kabiraj et al., 2018). In fact, this kind of wastes is commonly used as skeleton, given to  
691 their lower reactivity, since already fired, instead of hard components. The incorporation of fired

692 scraps into porcelain stoneware tiles (or other porcelain-like products) is quite described in the  
 693 literature, up to 20% in complete substitution of quartz or feldspars.  
 694 Residues generated during frit manufacturing are seldom recycled in ceramic tiles (García-Ten et  
 695 al., 2015). Their chemical composition is usually rich in alkalis, alkali-earths, and even strong fluxes,  
 696 like B and Zn (Table 8B).

Table 8A. Chemical composition of residues from ceramic production used in (or proposed for) ceramic tile manufacturing.

% wt.	Porcelain stoneware green scraps	Stoneware green scraps and sludge (4)			Sanitary-ware sludge	Glazing sludge (4)			Ceramic tile polishing sludge (7)			Filter dust
	Rambaldi et al., 2016	min	max	mean	Theerapap. & Nilpairach, 2015	min	max	mean	min	max	mean	El-Fadaly et al., 2010
SiO <sub>2</sub>	<b>69.81</b>	63.30	67.38	<b>65.87</b>	<b>57.26</b>	54.51	58.09	<b>56.76</b>	60.90	70.84	<b>65.96</b>	<b>64.38</b>
TiO <sub>2</sub>	<b>0.58</b>	0.65	0.98	<b>0.77</b>	<b>0.39</b>	0.09	0.16	<b>0.12</b>	0.15	0.48	<b>0.27</b>	<b>0.98</b>
ZrO <sub>2</sub>				<b>0.20</b>		2.47	11.90	<b>7.03</b>	1.10	2.51	<b>1.63</b>	
B <sub>2</sub> O <sub>3</sub>				<b>0.10</b>				<b>2.60</b>			<b>0.93</b>	
Al <sub>2</sub> O <sub>3</sub>	<b>18.85</b>	15.35	21.83	<b>18.22</b>	<b>27.77</b>	7.93	15.68	<b>11.54</b>	15.20	23.15	<b>18.75</b>	<b>19.42</b>
Fe <sub>2</sub> O <sub>3</sub>	<b>0.62</b>	2.10	4.25	<b>3.60</b>	<b>1.48</b>	0.14	0.52	<b>0.38</b>	0.37	1.41	<b>0.76</b>	<b>2.64</b>
MgO	<b>0.43</b>	0.79	2.99	<b>1.44</b>	<b>0.51</b>	0.71	2.41	<b>1.76</b>	2.21	4.63	<b>3.23</b>	<b>2.35</b>
CaO	<b>0.61</b>	2.57	3.00	<b>2.74</b>	<b>0.10</b>	6.05	9.70	<b>8.15</b>	0.62	3.81	<b>1.52</b>	<b>1.45</b>
MnO				<b>0.03</b>	<b>0.01</b>						<b>0.10</b>	<b>0.04</b>
ZnO				<b>0.20</b>		3.23	5.29	<b>4.39</b>			<b>0.65</b>	
SrO								<b>0.07</b>				
BaO				<b>0.10</b>		0.79	1.12	<b>0.97</b>			<b>0.65</b>	
PbO								<b>0.02</b>				
Na <sub>2</sub> O	<b>2.61</b>	1.80	2.75	<b>2.47</b>	<b>1.27</b>	1.37	5.49	<b>2.71</b>	1.79	5.17	<b>3.66</b>	<b>2.74</b>
K <sub>2</sub> O	<b>1.56</b>	1.43	2.80	<b>1.81</b>	<b>1.73</b>	1.36	3.54	<b>2.60</b>	1.19	3.40	<b>1.98</b>	<b>1.36</b>
P <sub>2</sub> O <sub>5</sub>				<b>0.19</b>	<b>0.04</b>			<b>0.06</b>				<b>0.50</b>
SO <sub>3</sub>	<b>0.09</b>			<b>0.14</b>				<b>0.06</b>			<b>0.09</b>	
Cl				<b>0.03</b>				<b>0.05</b>			<b>0.79</b>	
L.o.l.	<b>4.18</b>	4.00	6.00	<b>4.59</b>	<b>9.44</b>	1.70	3.22	<b>2.39</b>	1.97	40.03	<b>9.04</b>	<b>4.24</b>

Stoneware sludge: El-Fadaly et al., 2010; Garcia-Ten et al., 2015; Elmahgary et al., 2018; Amin et al., 2019. Glazing sludge: Wannagon et al., 2012; Dal Bò & Hotza, 2013; Nandi et al., 2015; Garcia-Ten et al., 2015. Ceramic tile polishing sludge: Rambaldi et al., 2007a; Xi et al., 2012; Silveira et al., 2014; Xian et al., 2015; Garcia-Ten et al., 2015; Ke et al., 2016.

697

698 *Sludge from ceramic tile polishing* is a byproduct of surface polishing operations aimed at improving  
 699 the tile aesthetics (providing brightness and mirror glass effect) and turning the final product similar  
 700 to natural stones. As a result, the polishing sludge, which is obtained by wastewater treatment to a  
 701 semisolid consistence, contains both ceramic dust and fragments of abrasive tools (Table 8A).  
 702 Usually, amounts of 1-5% of abrasive (SiC) and 2-6% of binder (magnesium oxychloride) are

703 declared (Shui et al., 2011) but there is a remarkable variability upon the surface treatment: from  
 704 deep polishing to mild lapping. The latter is currently carried out with diamond tools with a minimal  
 705 mass removal. Polishing sludge was tested as addition (up to 80%) to the mill, either in place of  
 706 feldspars or of the whole batch for porcelain stoneware tiles.

Table 8B. Chemical composition of residues from ceramic production used in (or proposed for) ceramic tile manufacturing.

% wt.	Porcelain stoneware fired scraps (93)			Stoneware fired scraps	Vitreous china scraps	Fireclay scraps	Sanitary-ware pitcher	Frit waste	Spent lime of kiln flue gas scrubber (7)		
	<i>min</i>	<i>max</i>	<b>mean</b>	Garcia-Ten et al., 2015	Tarhan et al., 2017	Tarhan et al., 2016	Fortuna et al., 2014	Garcia-Ten et al., 2015	<i>min</i>	<i>max</i>	<b>mean</b>
SiO <sub>2</sub>	58.7	80.0	71.4	67.2	68.58	74.29	70.8	60.6	0.32	1.93	1.10
TiO <sub>2</sub>	0.1	0.9	0.5	0.7	0.80	0.40	0.4	0.1			0.01
ZrO <sub>2</sub>	<0.1	1.0	0.8	0.2	0.29	0.19		2.9			0.01
B <sub>2</sub> O <sub>3</sub>				0.1				8.0			0.22
Al <sub>2</sub> O <sub>3</sub>	13.8	25.3	19.0	17.9	21.96	21.44	22.0	4.7	0.10	0.76	0.28
Cr <sub>2</sub> O <sub>3</sub>									0.15	0.29	0.22
Fe <sub>2</sub> O <sub>3</sub>	0.4	1.3	0.7	4.1	0.78	0.90	0.6	0.1	0.10	0.26	0.18
MgO	0.1	2.5	0.6	1.2	0.40	0.29	0.3	2.9	0.36	0.83	0.52
CaO	0.2	4.5	1.1	3.3	0.75	0.52	0.7	12.0	44.91	65.76	56.85
ZnO				0.3				6.6	0.12	0.37	0.20
BaO				0.1				2.4			0.02
PbO									0.22	12.87	8.35
Na <sub>2</sub> O	1.8	6.0	3.6	1.9	3.13	0.09	3.6	2.0	0.13	0.81	0.42
K <sub>2</sub> O	2.0	4.6	2.2	3.0	1.22	0.99	1.4	3.4	0.30	2.05	0.75
SO <sub>3</sub>									2.65	11.78	7.59
SeO <sub>3</sub>									0.48	0.64	0.56
F									26.77	32.60	30.95
Cl									0.32	1.60	1.20
L.o.l.				0.2	0.88	0.37	0.3	0.2	7.63	14.40	12.18

Porcelain stoneware fired scraps: Zanelli et al., 2011. Spent lime of kiln scrubber: Dondi et al., 1990; Andreola et al., 1992a, 1993; Garcia-Ten et al., 2015; Rambaldi et al., 2016.

707

708 *Spent lime produced by flue gas scrubbers* of roller kilns is a hazardous waste. During the firing  
 709 process, several volatile molecules are evolved, coming from both raw materials and additives.  
 710 Chlorine, fluorine and sulfur acids and compounds, which are released within the kiln, are subjected  
 711 to stringent regulations, which make mandatory the presence of filtration systems able to retain them.  
 712 These filters are based on powders or lumps of basic substances, generally lime, which selectively  
 713 react with some contaminants present in flue gases, thus blocking them. In fact, the chemical

714 composition of spent lime is mainly constituted, along with Ca (calcite and calcium hydroxide), by a  
715 large percentage of fluorine (mostly as calcium fluoride) and, in smaller amounts, by chlorine and  
716 sulfur, plus ingredients of glazes and pigments (B, Cr, Se, Zn). At any rate, concentrations are  
717 strongly affected by the specific ceramic production: the occurrence of PbO, for instance, was  
718 registered in the analyses pre-2000 ([Table 8B](#)). Since this waste is currently landfilled, its recycling  
719 within the same tile factory would represent an obvious pathway. As the waste occurs in the form of  
720 powder, it can be easily introduced in the milling step ([Andreola et al., 1992a, 1992b](#)). This recycling  
721 solution was practiced during 1970s and 1980s, adding the waste to ceramic batches fired at  
722 relatively low temperatures ([Andreola et al., 1993](#)). Nonetheless, it was gradually abandoned with  
723 the conversion to porcelain stoneware, when it emerged that fluorine is released at temperatures  
724 >1000°C, depending on firing cycle ([Dondi et al., 1990](#)). In addition, chlorine can be even more easily  
725 released during the firing process, increasing its content into vent streams. For these reasons, the  
726 recent efforts to include the spent lime in optimized batches, designed to get 100% waste porcelain  
727 stoneware tiles ([Garcia-Ten et al., 2015; Rambaldi et al., 2016, 2018](#)) had no industrial repercussion.

#### 728 *4.8. Wastes from construction, demolition, and dredging*

729 These waste sources attracted a limited attention as potential supply of the ceramic tile production.  
730 Most research efforts were addressed to recycling of sediments from dredging harbors, rivers, and  
731 estuaries (often contaminated) or construction and demolition wastes (CDW), particularly asbestos-  
732 containing residues. Interestingly, research was carried out not only at the laboratory level, but the  
733 scale-up to a commercial secondary raw material was achieved in the case of CDW.

734 *Construction and demolition wastes* include common residues – i.e., mixture of brick, stone,  
735 concrete, and mortars in variable ratios ([Table 4](#)) – that were experimented by mixing with a red clay  
736 ([Acchar et al., 2013b](#)). The outstanding variability of CDW in both composition and particle size  
737 distribution ([Bianchini et al., 2005; Rodrigues et al., 2013](#)) and the possible mobilization of heavy  
738 metals, such as Cr, Se, Mo, Cu, S, Cl ([Delay et al., 2007; Butera et al., 2014](#)) represent obstacles  
739 for their recycle. In order to overcome this hindrance, the importance of selective waste collection is  
740 stressed ([Acchar et al., 2013b](#)). This is the starting point in the treatment of hazardous demolition  
741 residues, essentially cement-asbestos slates ([Leonelli et al., 2006; Gualtieri et al., 2008](#)). Given the  
742 known carcinogenic hazard, asbestos must be completely transformed into inert silicate phases, e.g.

743 by melting to a silicate glass at  $T > 1250^{\circ}\text{C}$  (Gualtieri et al., 2008) or by microwave inertization  
744 (Leonelli et al., 2006). By such preliminary treatment, asbestos was transformed into harmless  
745 forsterite. Laboratory trials with porcelain stoneware batches containing the transformed asbestos  
746 residue (5%), fostered the scale up to the industrial production of a secondary raw material (KRY-AS)  
747 obtained by the thermal transformation of cement-asbestos into properly designed glass-ceramic  
748 frits (Table 4). Cement kiln dust is a fine-grained residue rich in CaO, silica and alumina (Table 4)  
749 that was tested (up to 15%) in a wall tile batch in substitution of calcite (Aydin et al., 2019).

750 *Dredging soils* resulting from river or harbor dredging are sediments with a composition approaching  
751 that of clay raw materials, more or less rich in sand or carbonate fractions. Nonetheless, they are  
752 considered hazardous materials because of possible contamination by heavy metals, such as As,  
753 Cd, Cr, Cu, Pb, V and Zn (Tack et al., 1999; Borma et al., 2003) as well as organic substances (e.g.,  
754 hydrocarbons). Relevant from the technological point of view are the relatively high amount of iron  
755 oxide and moisture (semi-solid wastes). On the other hand, the modest concentrations of S and Cl  
756 may be matter of concern for volatile emissions. Similarly, sludges stemming from purification of  
757 drinking water often have a composition close to that of clay sediments, particularly rich in aluminium  
758 and iron oxides (Table 9). Dredging spoils were investigated as ingredients to produce ceramic tiles:  
759 alone (Baruzzo et al., 2006; Romero et al., 2008) or up to 20% in replacement of quartz (Hossain et  
760 al., 2019) or clay materials (Torres et al., 2009). Analogous trials were run using the sediment (up to  
761 65%) from water supply treatment (Junkes et al., 2012; Wangrakdiskul et al., 2017; Rodrigues and  
762 Holanda, 2018).

#### 763 4.9. Wastes from municipal wastewater treatment plants

764 Municipal sewage sludges (MSS) are obtained from water treatment plants residues, as many types  
765 as the different process stages (Świerczek et al., 2018). Nowadays, MSS handling is a real problem  
766 and their utilization in ceramic production has been extensively studied (Chang et al., 2020). The  
767 main issue is its water content up to 95-98% that turns into a semi-liquid waste (Chang et al., 2020).  
768 For this reason, MSS needs a previously drying that requires a high energy consumption, limiting  
769 any application at industrial scale. MSS composition is generally quite variable, depending on various  
770 factors which affect amount and nature of the inorganic and organic sludge compounds (Sommers,

771 [1977; Dondi et al., 1997](#)). Furthermore, the purifying process entails the use of ferric or aluminum  
 772 chloride as flocculant that joins the presence of toxic elements ([Xu et al., 2013; Yang et al., 2013](#)).  
 773 Potential biohazards, i.e. microorganism, pathogens, are known ([Giger et al., 1984; Giller et al.,](#)  
 774 [1998; Gerba and Smith, 2005; Ruan et al., 2012](#)) as problems with odors ([Cremades et al., 2015,](#)  
 775 [2018](#)). Sewage sludges tested in ceramic tiles exhibit significant concentrations of heavy metals,  
 776 namely Cr, Co, Cu, Ni, Sb, Sn and Zn ([Carlson and Morrison, 1992; Fjällborg et al., 2005](#)) along with  
 777 rather high amounts of P, S and Cl ([Table 9](#)).

Table 9. Chemical composition of residues from wastewater treatment and dredging operations used in (or proposed for) ceramic manufacturing.

% wt.	Sewage sludge (8)			Dredging spoils, harbour sediments (5)			Drinking water purification sludge (16)		
	<i>min</i>	<i>max</i>	<b>mean</b>	<i>min</i>	<i>max</i>	<b>mean</b>	<i>min</i>	<i>max</i>	<b>mean</b>
SiO <sub>2</sub>	9.46	38.01	<b>24.18</b>	47.13	57.38	<b>52.82</b>	10.90	60.85	<b>40.93</b>
TiO <sub>2</sub>	0.26	1.25	<b>0.75</b>	0.55	0.84	<b>0.65</b>	0.16	1.28	<b>0.79</b>
Al <sub>2</sub> O <sub>3</sub>	2.62	13.75	<b>9.02</b>	7.69	20.29	<b>12.33</b>	1.34	62.66	<b>26.38</b>
Cr <sub>2</sub> O <sub>3</sub>	0.02	0.04	<b>0.03</b>			<b>0.03</b>			
Fe <sub>2</sub> O <sub>3</sub>	4.26	16.70	<b>7.74</b>	3.79	21.55	<b>8.11</b>	1.16	68.65	<b>11.73</b>
As <sub>2</sub> O <sub>3</sub>						<b>0.04</b>			
MgO	0.87	2.55	<b>1.60</b>	1.46	25.16	<b>6.98</b>	0.15	3.08	<b>1.17</b>
CaO	3.97	42.46	<b>15.58</b>	0.69	7.48	<b>4.33</b>	0.09	11.15	<b>2.71</b>
MnO			<b>0.03</b>	0.03	0.31	<b>0.10</b>			
CuO	0.07	0.27	<b>0.17</b>			<b>0.01</b>			
ZnO	0.31	0.44	<b>0.36</b>			<b>0.03</b>			
SrO						<b>0.02</b>			
Na <sub>2</sub> O	0.31	1.23	<b>0.67</b>	0.42	2.00	<b>1.25</b>	0.02	1.53	<b>0.46</b>
K <sub>2</sub> O	0.53	2.37	<b>1.51</b>	0.01	3.37	<b>1.73</b>	0.18	3.62	<b>1.53</b>
P <sub>2</sub> O <sub>5</sub>	3.81	28.36	<b>15.74</b>	0.09	0.23	<b>0.17</b>	0.17	5.89	<b>1.29</b>
V <sub>2</sub> O <sub>5</sub>						<b>0.30</b>			
SO <sub>3</sub>	3.31	13.34	<b>6.42</b>			<b>0.86</b>			
Cl	0.08	0.27	<b>0.18</b>			<b>0.10</b>			
L.o.l.	32.09	66.12	<b>50.12</b>	6.90	19.35	<b>13.74</b>	5.11	39.77	<b>19.69</b>

Sewage sludge: [Jordan et al., 2005; Montero et al., 2009b; Jordan, 2010; Zhou et al., 2013; Amin et al., 2018](#). Dredging spoil: [Baruzzo et al., 2006; Romero et al., 2008; Torres et al., 2009](#). Drinking water sludge: [Junkes et al., 2012; Wangrakdiskul et al., 2016; Rodrigues and Holanda, 2018; Sarabia et al., 2019](#).

778

779 Few studies considered the possibility to use MSS in tiles production, even if a wide range of  
 780 concentration (1-65%) was studied ([Abadir et al., 2004; Jordán et al., 2005; Montero et al., 2009b;](#)  
 781 [Jordán, 2010; Amin et al., 2018; Chang et al., 2020](#)).



#### 782 4.10. Solid wastes from municipal and industrial sorting

783 Solid wastes from municipal and industrial sorting include different types of glass. They derive from  
784 the separated collected fractions of: container and flat glass, referred as Soda-Lime-Silica (SLS);  
785 glasses from Waste Electrical and Electronic Equipment (WEEE): PC-TV screen, PC-TV cathodic  
786 tube (CRT), different types of lamps; other post-consumer glasses (such as thin-film transistor liquid-  
787 crystal display TFT-LCD, veal, solar panels); glasses from waste melting (municipal solid waste  
788 incineration, dredging spoils, vitrification of various wastes). All are amorphous materials, or present  
789 a negligible crystalline component, and can be basically distinguished on their chemical composition  
790 (Table 10). The main issue, which prevents any direct use in the ceramic tile process, is the glass  
791 particle size, which depends on the waste collection system and following treatments. A  
792 technological gap usually exists and imposes a milling before use as ceramic raw materials. Other  
793 pre-treatments are related to the presence of pollutants: e.g., labels and glue (causing rheological  
794 problems to the slips) or residues of plastics, metal circuits, or functional coatings that must be  
795 removed. Some concerns on environmental and health problems are related to the presence of Pb,  
796 Sr and Ba in cathode-ray tube and panel waste glass (Musson et al., 2000; Méar et al., 2006; Yot  
797 and Méar, 2011); As and Sb in LCD (Savvilotidou et al., 2014); Hg in fluorescent lamp waste (Rey-  
798 Raap and Gallardo, 2012; Tunsu et al., 2014). They have to be taken into account, particularly in  
799 case of obsolete materials, which no longer respond to the current law requirements.

800 The use of waste glasses in the ceramic tile production has been extensively investigated in the last  
801 decades, for their fluxing power able to lower the firing temperature (Silva et al., 2017; Raimondo et  
802 al., 2007). Especially, the employ of SLS in substitution of feldspars in porcelain stoneware was the  
803 object of many papers. SLS has been also evaluated in dark-firing bodies (vitrified, porous and split  
804 tiles), and waste-based bodies too, providing a variegated picture regarding the employment of this  
805 type of glass. However, a large part of literature used lab scale conditions far from those of the  
806 ceramic industry, with slow firing cycles. Anyway, some example of industrial-like cycle at lab scale  
807 (Matteucci et al., 2002; Pontikes et al., 2005; Faria and Holanda, 2016), tests on a pilot line  
808 (Carbonchi et al., 2003; Tucci et al., 2004) and even a prototype production (Skerratt, 2001), were  
809 found. As concern the other glasses, their use was much less examined.

Table 10. Chemical composition of waste glasses from municipal and industrial sorting used in (or proposed for) ceramic tile manufacturing.

% wt.	Soda-lime glass container & flat (34)			PC-TV screen glass (9)			TV cathode tube glass (7)			Lamp glasses				Other post-consumer glasses				Waste vitrification			
	min	max	mean	min	max	mean	min	max	mean	Bulb	Neon	Fluorescent	Mixed	Veal	LCD	Solar panel	MSW ash	Dredge spoils	Tailings+lime+SLS		
SiO <sub>2</sub>	64.83	77.07	71.67	58.90	66.05	62.13	50.35	56.72	53.17	84.79	70.06	64.95	75.92	73.50	60.10	72.33	40.70	46.08	40.70		
TiO <sub>2</sub>	0.01	0.20	0.08	0.03	0.43	0.31	0.08	0.19	0.12	0.08	0.07	0.00		1.00			1.50	0.72	1.50		
ZrO <sub>2</sub>	0.01	0.17	0.08	0.01	2.90	1.07	0.20	0.85	0.41				0.48	11.00	10.30						
B <sub>2</sub> O <sub>3</sub>						0.20	0.10	0.50	0.25				2.20	6.00	16.80		12.60	12.55	12.60		
Al <sub>2</sub> O <sub>3</sub>	0.01	6.02	1.64	1.80	4.38	2.98	2.30	4.10	3.25	2.98	2.14	4.31	2.20	6.00	16.80	1.90	12.60	12.55	12.60		
Cr <sub>2</sub> O <sub>3</sub>										0.03	0.02						0.70		0.70		
Fe <sub>2</sub> O <sub>3</sub>	0.10	0.86	0.32	0.07	0.80	0.21	0.08	0.20	0.12	0.15	0.17	0.10	0.05	0.50	0.03	0.03	1.00	6.24	1.00		
MgO	0.12	4.43	2.80	<0.01	1.20	0.51	1.30	2.02	1.76	0.28	3.25		1.85	0.00	0.44	2.62	3.30	6.30	3.30		
CaO	7.45	24.84	10.08	<0.01	2.20	0.82	2.70	3.86	3.40	1.56	6.66	5.26	3.90	0.75	7.60	8.98	32.20	20.00	32.20		
MnO	0.01	0.05	0.02						0.01	0.02	0.07										
NiO				0.03	0.32	0.11	0.02	0.34	0.18												
CuO																					
ZnO			0.01	<0.01	0.50	0.20	0.09	0.22	0.13												
SiO			0.02	0.94	9.60	5.71	0.70	3.05	1.45						4.20						
BaO	0.02	0.19	0.09	8.40	12.00	10.55	0.80	4.03	2.39					1.00	0.48						
PbO	0.08	0.50	0.17	<0.01	0.40	0.15	15.58	24.60	21.09												
Na <sub>2</sub> O	0.47	16.48	12.34	7.05	8.60	7.82	6.10	7.00	6.61	4.62	14.77	19.53	12.45	6.25	12.87		4.60	3.04	4.60		
K <sub>2</sub> O	0.07	4.65	0.74	5.56	7.10	6.15	5.37	7.70	6.77	1.68	0.69	1.10	3.15	0.75	0.04		0.30	2.80	0.30		
P <sub>2</sub> O <sub>5</sub>	0.01	0.19	0.05				<0.01	0.02	0.01	0.43	1.66						2.20		2.20		
Sb <sub>2</sub> O <sub>5</sub>				0.10	0.46	0.38	<0.01	0.66	0.30												
SO <sub>3</sub>			0.20							0.07	0.07										

Soda-lime glass: Youssef et al., 1998; Matteucci et al., 2002; Carbonchi et al., 2003; Rambaldi et al., 2004; Souza et al., 2004; Bakr, 2005; Pontikes et al., 2005; Tarvorpanich et al., 2005; Tucci et al., 2006; Luz et al., 2007; Pontikes et al., 2007b; Rambaldi et al., 2007b; Bernardo et al., 2009; Maschio et al., 2009; Viruthagiri et al., 2009; Yürüyen et al., 2009; Bernardo et al., 2011; Fraga et al., 2011; Mustafi et al., 2012; Wannagon et al., 2012; Marinoni et al., 2013; Fortuna et al., 2014; Ajanaku et al., 2015; Theerapapisetpong et al., 2015; Faria and Holanda, 2016; Rambaldi et al., 2016; Barrachina et al., 2018; Chitwaree et al., 2018; Gualtieri et al., 2018; Njindam et al., 2018; Darweesh, 2019. PC-TV screen: Andreola et al., 2005; Tucci et al., 2006; Raimondo et al., 2007; Andreola et al., 2008; Bernardo et al., 2008b; Bernardo et al., 2009; Andreola et al., 2010; Revelo et al., 2018. TV cathode tube CRT: Rambaldi et al., 2004; Souza et al., 2004; Andreola et al., 2005; Tucci et al., 2006; Raimondo et al., 2007; Bernardo et al., 2009; Nandi et al., 2015.

811 *Soda-Lime-Silica glass* is characterized by a homogeneous chemical composition (Table 10). At  
812 variance of fluxes utilized in the ceramic tile industry, such as feldspars, SLS shows lower contents  
813 of alumina and higher levels of soda and lime, resulting more fusible. For this reason, SLS has been  
814 mainly employed as flux in substitution of feldspars (up to 30%) in porcelain stoneware batches  
815 (Matteucci et al., 2002; Carbonchi et al., 2003; Rambaldi et al., 2004, 2007b; Tucci et al., 2004, 2006;  
816 Bakr, 2005; Pontikes et al., 2005; Tarvornpanich et al., 2005; Mustafi et al., 2012; Ajanaku et al.,  
817 2015). Nevertheless, other strategies of incorporation were tested, e.g., in substitution of clay (up to  
818 50%) mostly for red stoneware (Theerapapvisetpong et al., 2015; Njindam et al., 2018), with one  
819 example up to 5% for porous tiles (Faria and Holanda, 2016). Otherwise, trials were run with glass  
820 in replacement of the whole body (up to 50%) in red stoneware (Youssef et al., 1998), porous  
821 (Youssef et al., 1998; Darweesh, 2019) or split tiles (Pontikes et al., 2007b). Waste-based products  
822 were also investigated, such as SLS mixed with cockle shells in proportion from 50:50 to 70:30  
823 (Shamsudin et al., 2018) or 95% SLS plus additives (Skerratt, 2001).

824 *PC-TV screen glass* represents the front part of television and computer monitors with a chemical  
825 composition enriched in alkali ( $\text{Na}_2\text{O}+\text{K}_2\text{O} = 14\%$ ), barium and usually strontium (Table 10). Barium  
826 released by leaching from the as-received waste glass is in very low amounts (Andreola et al., 2008).  
827 PC-TV glass has been investigated in substitution of feldspars (up to 35%) in porcelain stoneware  
828 (Tucci et al., 2006; Raimondo et al., 2007, Andreola et al., 2008) and in place of clays (up to 20%)  
829 in red stoneware (Andreola et al., 2010).

830 *PC-TV cathodic tube* is the hidden part inside the PC-TV set composed of a low melting temperature  
831 glass, showing lower levels of silica, comparable alkali and very high content of Pb, with respect to  
832 both SLS and PC-TV screen glasses (Table 10). For this reason, CRT has been investigated as flux  
833 in substitution of feldspars in porcelain stoneware (up to 10%) sometimes coupled with SLS up to  
834 5% (Rambaldi et al., 2004; Tucci et al., 2006; Raimondo et al., 2007).

835 *Lamp glass* includes different types of waste glass (Table 10): from incandescence bulb,  
836 characterized by a high silica content, as well as from neon (Chemani, 2012) and fluorescent lamps  
837 (Morais et al., 2011; Araùjo et al., 2014; Andreola et al., 2016) particularly rich in sodium. Even mixed  
838 lamps were investigated (Gaitanelis et al., 2018). Such a heterogeneity in composition represents a  
839 challenge for recycling because the different waste lamps are generally recovered all together. In

840 addition, these mixtures of various glasses vary randomly, according to the changing customs.  
841 Significant technological data are given only for glasses from incandescence (Chemani, 2012) and  
842 fluorescent lamps (Morais et al., 2011) used in substitution of feldspars (up to 15%) and clays (up to  
843 10%) in porous tiles.

844 *Other post-consumer glasses* stem from solar panels, substantially similar to SLS (Lin et al., 2015),  
845 as well as TFT-LCD (Lin, 2007; Kim et al., 2015, 2016) and veal (Gualtieri et al., 2018) that are both  
846 rich in boron and alumina, plus Ca-Sr in LCD and Na in veal (Table 10). Unlike LCD produced before  
847 2010, the current glasses do not contain toxic components, such as arsenic and antimony oxide  
848 (Kim et al., 2015). Only two papers report technological results on LCD glass in replacement of  
849 feldspars (up to 40%) in porcelain stoneware (Kim et al., 2016) or in place of clays (up to 50%) in  
850 porous tiles (Lin, 2007).

851 *Glasses from waste melting* derive from different vitrification processes: municipal solid waste  
852 incinerator fly ashes by the application of thermal plasma melting (Bernardo et al., 2011); polluted  
853 dredging spoils from industrial area (Brusatin et al., 2005); feldspar mining residues and lime from  
854 fume abatement system of the glass industry, admixed with SLS (Bernardo et al., 2008a, 2008b) or  
855 PC-TV glass (Bernardo et al., 2008b); air pollution control residues from thermal energy plants  
856 (Deveraj et al., 2010). Overall, these wastes have a low percentage of silica but are rich in alumina  
857 and particularly CaO (Table 10). Air cleaning residues have a problematically high chlorine (~2%).  
858 The glass obtained by plasma melting (Deveraj et al., 2010; Bernardo et al., 2011), as well as those  
859 from mining residues vitrified with lime and SLS (Bernardo et al., 2008a), were used in place of the  
860 non-plastic component (up to 70%) in a novel kind of “glass-ceramic stoneware”. Otherwise, the  
861 waste derived from a mixture with PC-TV glass was used in substitution of feldspars (up to 7%) in a  
862 porcelain stoneware batch (Bernardo et al., 2008b). Vitrified dredging spoils were used in  
863 replacement of clay (up to 10%) in red stoneware (Brusatin et al., 2005).

## 864 **5. Technological feasibility of waste recycling in tile-making**

865 In the previous chapters (and supplementary material), the effect of various kinds of waste was  
866 described to point out the feasibility of recycling from technological and environmental viewpoints.  
867 This assessment is here summarized, waste by waste, attributing a Technology Readiness Level to

868 both recycling solutions proposed in the literature and known industrial achievements (Fig. 6). Such  
869 a degree of development goes from TRL 4 (waste recycling evaluated at the laboratory scale with  
870 processing conditions far from the industrial production) to TRL 9 (technological solution  
871 industrialized and put on the market). In any case, some caution must be used in taking the TRL  
872 indicated in Figure 6, because it depends on both production technology and type of tile. A waste  
873 that causes intolerable interferences to the slip rheological properties, for instance, might be utilized  
874 in the production of tiles by the dry route. Furthermore, a waste that induces an increase in porosity,  
875 unacceptable for porcelain stoneware tiles, could instead be used in semi-vitrified products, which  
876 have a less stringent threshold in terms of water absorption. The same applies to the mechanical  
877 properties of finished and semi-finished products, and even more to the aesthetic appearance: a  
878 darkening of the body color, hardly tolerated in unglazed porcelain stoneware, could be passable for  
879 other types of tile. Ultimately, we were conservative in the evaluation of the TRL, which was  
880 estimated mostly with reference to porcelain stoneware tiles, since it is the most studied type, and  
881 which offers more options for recycling waste.

882 The ranking in Figure 6 highlights some cases of waste recycling that since many years entered in  
883 the industrial practice (TRL 9). Relevant examples include: granite blocks recovered from quarry  
884 dumps and beneficiated (Dino et al., 2012); glass cullet from sorting of municipal solid wastes  
885 (Carbonchi et al., 2003); ceramic residues, such as sludges, unfired tiles, dust, and fired scraps  
886 (Manfredini et al., 1991; García-Ten et al., 2015; Rambaldi et al., 2018). These recycling operations  
887 led either to commercial fluxes (feldspathic materials, also improved with soda-lime waste glass) or  
888 to in-house byproducts, usually cannibalised in the tile-making process (introducing them in the  
889 milling stage). At present, such cases of successful recycling regard wastes with composition and  
890 technological behavior that are close to those of ceramic raw materials. This rule of thumb can be  
891 extended to other cases, although the lesser is the chemical affinity with ceramic tile batches, the  
892 lower is the recyclable amount (e.g., glass cullet <5% versus granite blocks up to 20%).

Source ▼	Property ►	technological feasibility (TRL)	before the tile factory			body preparation and shaping				firing and tile performances				environmental impact		
			crushing, milling, deagglomeration	dewatering, drying	handling, storage, stabilization	grindability, slip particle size	rheological behavior of slip and powders	compressibility, green bulk density	unfired bending strength, springback	shrinkage, overfiring, pyroplasticity	bulk density, porosity, water absorption	bending strength, abrasion resistance	color and defects	leaching of hazardous elements	VOC release, bad smell	inorganic gaseous emissions
Excavation/cleaning of minerals	9	●	●		○	○	○	○				○				
Tailings from ore processing	5		●	●		●		●	●	○		●	●		●	
Tailings from borate processing	5		●	●		●			●	○		●	○		●	
Marble cutting and sawing sludge	8		●	○		●	○	○		●	●					
Granite cutting and sawing sludge	6		●	○		●	○	○	●	●		●				
Pulp and paper industry	4	○	●	●		●	○	○		●	●		●	○	○	
Petroleum and edible oil refining	5		●	●	○	●			●	○			○	●	○	
Titania sludge	5		○			●	○	○	●	○		●	○		●	
Alumina red mud	4		●	●		●	○	○	●	○		●	○			
Coal fly ash	6			●		●	●	●	○	○		●	○	○	●	
Coal bottom ash	5	●		●	○	●				○		●	○	○	●	
MSWI bottom ash	8	●		●	○	●				○		●	○	○	●	
MSWI fly ash	4			●		●	●	●		○		●	○	○	●	
Sewage sludge ash	4			●		●	○			○	●	○	○	○	●	
Biomass fly and bottom ashes	5	●	○	●	○	●	○	○	●	○		●		○	●	
Blast furnace slag	5	●			○		○	○				○				
Electric arc furnace slag	5	●			●		○	○	○		○	●	○			
Basic oxygen furnace slag	4	●			●		○	○	○		○	●	○			
Non-ferrous slag	5	●			●		○	○	○		○	●	○			
Spent foundry sand	6	○			○	●	○	○	○		○	○	○	○	○	
Galvanic sludge	5		●	○		●	○	○	●			●	●		●	
Ceramic unfired waste	9	○														
Ceramic fired scrap	9	●			○		○	○								
Ceramic wastewater sludge	9		○	○		○						○			○	
Ceramic polishing sludge	7		○			○	○	○	○	●		○			○	
Spent lime of kiln scrubber	8			○		●			○	○		○	○		●	
Construction and demolition waste	4	●				○	○	○	○	○		○	○		○	
Transformed cement-asbestos	6	●		●	○	○	○	○	○	○		○	○	●		
Harbor and river dredging	5		●	●		○			○			○	●		○	
Drinking water sludge	5		●	○		○			○			○		○		
Sewage sludge	6	○	●	●		○	○		○	○	○	○	○	○	○	
SIS container glass	9	●			○		○	○	○	○		○				
PC-TV screen glass	8	●			○		○	○	○	○		○				
PC-TV cathode tube glass	6	●			○		○	○	○	○		○	○	○		
lamp glass	8	●			○		○	○	○	○		○				
Further glasses from WEEE	4	●			○		○	○	○	○		○			○	

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Figure 6. Technological feasibility of waste recycling in tile-making (TRL) and matters of concern due to the occurrence of waste along the ceramic manufacturing chain. Symbols refer to issues that can be critical, beyond the usual concern in tile-making, or manageable with approach and facilities of tile-makers.

897 A step below are technological solutions ready for ceramic tile production but not yet become an  
898 industrial reality (TRL 8). Therefore, waste recycling appears to be fully feasible, as in the case of  
899 PC-TV screen glass (Tucci et al., 2006; Raimondo et al., 2007), MSWI bottom ash (Andreola et al.,  
900 2001), lamp glasses or the exhausted lime from kiln scrubbers (García-Ten et al., 2015; Rambaldi  
901 et al., 2018). We are dealing with products already proposed on the market as secondary raw  
902 materials (or recycled in the same tile manufacturing) but at best of our knowledge not durably  
903 entered in the ceramic production. This is commonly due to technological hindrances that still remain  
904 unresolved. The misstep may depend on various causes: a supply gap (need of further beneficiation  
905 for mixed lamp glass, if still contaminated by plastic or metallic parts) or difficulties in batch design  
906 due to the exotic composition (high percentages of Ba and Sr of the screen glass) or release of  
907 harmful substances during firing (as for the spent lime and untreated bottom ashes).

908 The technological feasibility of recycling was proven in industrially relevant environment by trials at  
909 the pilot or the full scale for a wide range of wastes (TRL 6 to 7). The list includes: ornamental stone  
910 cutting and sawing sludge (Souza et al., 2010b; Silva et al., 2011), coal fly ash (Olgun et al., 2005;  
911 Kockal, 2011), spent foundry sand (Coronado et al., 2015; Lin et al., 2018), ceramic tile polishing  
912 sludge (Rambaldi et al., 2007a; Kabiraj et al., 2018), asbestos-containing demolition wastes (Ligabue  
913 et al., 2020), sewage sludge (Montero et al., 2009b; Amin et al., 2018), PC-TV cathode tube glass  
914 (Tucci et al., 2006; Raimondo et al., 2007). Nevertheless, the technological transfer has been  
915 somehow frozen before industrialization by obstacles to recycling. For instance, the slip rheological  
916 behavior, and consequently grindability, can be heavily affected by the spent foundry sand (if the  
917 bentonite binder is still active) or sludges stemming from sawing or polishing operations (because of  
918 soluble salts). The characteristics of unfired tiles can be worsened by coal fly ash and spent foundry  
919 sand (as their particles have a poor compressibility). Firing behavior can be strongly modified by  
920 cathode tube glass (being extremely fusible) or by sawing and polishing sludges, which foster  
921 bloating phenomena (due to SiC and/or iron oxide). It is the occurrence of transition elements,  
922 inherited from polished rocks and tiles, which determines a color darkening. But even more than  
923 interferences in the ceramic process, the need for preventive treatment and storage problems act as  
924 a brake, as in the case of sewage sludge and coal fly ash. Last but not least, the occurrence of

925 hazardous elements and the release of harmful substances during firing are a matter of concern,  
926 particularly about asbestos-containing demolition wastes, cathode tube glass and coal fly ash.

927 The remaining wastes were evaluated just at the laboratory scale. In some cases, the technological  
928 feasibility was assessed by an industrial-like processing (TRL 5) that makes the results promising  
929 for a scale up. Examples encompass mining tailings, oil refining sludge, titania sludge, coal bottom  
930 ash, biomass ashes, metallurgical slags, galvanic sludge, dredging soil (Fig. 6). In these cases, the  
931 criticalities seem to concern more the waste pre-treatment and environmental issues than the  
932 ceramic manufacturing. In fact, the supply gap appears to be severely limiting about: oil refining  
933 sludge, dredging soil and galvanic sludge (storage and handling problems for bad smell,  
934 fermentation, acid release); coal and biomass bottom ashes (moisture susceptible, tendency to  
935 cementation); metallurgical slags (needing to be crushed to few millimeters). Possible leaching of  
936 hazardous substances or harmful gaseous emissions during firing are a common risk for almost all  
937 these wastes. These issues are especially critical for galvanic sludge and dredging soil, even though  
938 they are the bottleneck also for coal and biomass bottom ashes, titania sludge and some mining  
939 tailings. The main limitations in the ceramic process are focused on the slip rheological properties  
940 (in case of ashes and sludges) or consequent to grindability and mechanical strength matters for  
941 hard wastes, like metallurgical slags. The remarkably high concentration in iron and other transition  
942 elements of some wastes (steel slags, galvanic and titania sludges) constitutes a divide between  
943 light-firing and dark-firing bodies that is difficult to circumvent, despite the fact that literature focuses  
944 on porcelain stoneware.

945 About the wastes investigated at TRL 4, the available information is often insufficient to conclude  
946 about technological feasibility of their recycling in the production of ceramic tiles. Further  
947 investigation is needed to achieve a reliable assessment in conditions closer to industrial  
948 manufacturing.

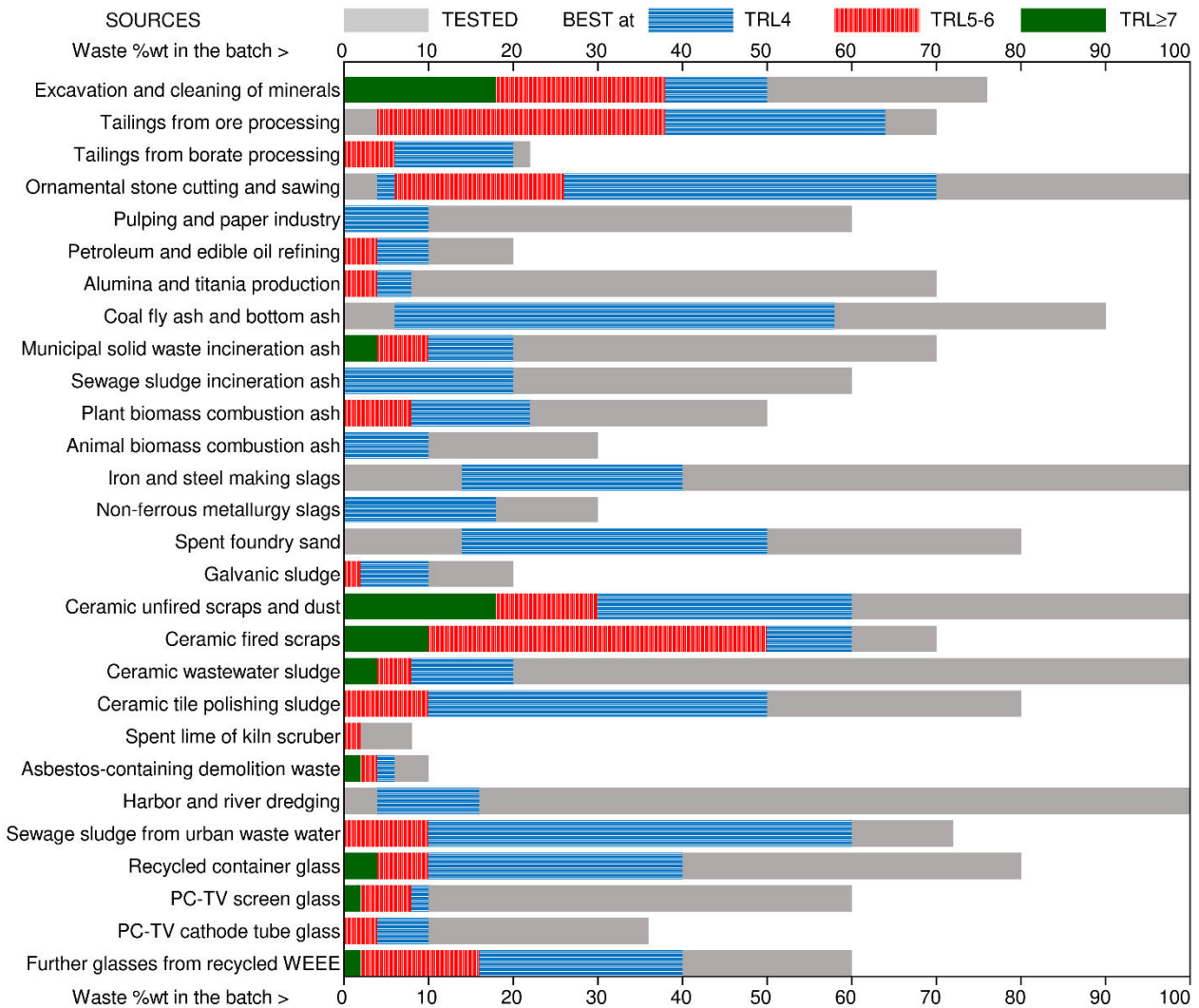
949 Looking in detail at Figure 6, it can be appreciated that the overall frequency of concerns is roughly  
950 the same by comparing the stages of ceramic production versus the aspects of pre-treatment plus  
951 environmental impact. Interestingly, the critical issues concerning the supply gap are more than  
952 those regarding the firing behavior. The same happens when comparing the stages of the ceramic  
953 process: the serious criticalities are more in the body preparation, shaping and drying phases than



954 during firing. This picture leads to a first consideration: the scientific literature, which is focused  
955 essentially on the firing stage, does not fully reflect the waste recycling priorities from an industrial  
956 point of view. This is probably due to the weight attributed to aspects that are often considered of  
957 little interest to technological research (i.e., to faithfully replicate industrial processes since the supply  
958 of raw materials). Another reason may be that many critical points relate to environmental issues,  
959 which are often studied separately. Nonetheless, by focusing on the ceramic process, it is clear that  
960 the most critical aspects concern two passages that are little considered in the literature: the  
961 rheological behavior of the slips and the aesthetic appearance of the finished product. This is  
962 perhaps dictated by the viewing angle of our evaluation, focused on the wet processing route and  
963 on light-firing tile bodies.

964 The question of the recommended quantity of waste, which means the maximum recyclable amount  
965 without having intolerable effects on the technological behavior and technical performance of  
966 ceramic tiles, was touched in the supplementary material. Such a recommended recyclable amount  
967 is summarized in [Figure 7](#), waste by waste, taking care to distinguish the indications drawn from  
968 different TRL. In fact, it is known that the recommended recyclable amount decreases with increasing  
969 TRL. This is a natural consequence of taking more and more technological, logistical, environmental,  
970 and economic aspects into consideration. To the rule of thumb, it can be said that the recommended  
971 percentages for  $TRL > 7$  are already recycled in industrial production. In reality, this never happens  
972 systematically in all ceramic factories in the world but indicates that it is certainly feasible for those  
973 industries equipped with the best available technologies.

974 Significantly, the percentage of waste recommended by the studies at TRL 5-6 overestimates the  
975 quantity that can be profitably recycled 2 to 4 times, depending on different factors (type of tiles,  
976 batch design strategies, and so on). On the other hand, TRL 4 experiments can give rise to  
977 overestimations of 3 to 10 times of the recyclable amount of waste in industrial conditions. These  
978 observations suggest extreme caution in affirming technological feasibility on the basis of results  
979 obtained at the laboratory scale. As an indication, the optimal waste quantity, based on TRL 5 trials,  
980 should prudently be halved at the time of giving a recommended percentage to ceramic tile industry.  
981 Data deriving from preliminary tests (TRL 4) should only serve to direct subsequent tests to higher  
982 TRLs, without general conclusions regarding the technological feasibility of waste recycling.



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Figure 7. Recommended recyclable amount for various wastes, according to tests carried out at the laboratory scale (TRL4, i.e. conditions not of industrial tile-making) or up to pilot scale (TRL5-6, i.e. conditions simulating industrial tile-making) or up to industrial manufacturing (TRL≥7).

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## 6. Conclusions

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The literature on waste recycling in ceramic tiles covers a range of products, from vitrified to porous, and different incorporation strategies, as replacement of raw materials in dark-firing and light-firing batches or in novel, waste-based bodies. This critical review addresses the problem of waste recycling from an industrial point of view, highlighting benefits but also constraints, according to a two-level approach: a general overview of the specific recycling problems of each residue, and a thorough analysis of technological behavior (reported in the supplementary material).

Since the main obstacle to a widespread recourse to waste recycling is the lack of knowledge about the effect on technological behavior and technical performance of ceramic tiles, these aspects have

996 been examined in detail, in order to outlook feasibility, recyclable amount, and possible matters of  
997 concern for every source.

998 The examined literature mostly focused on the firing step, despite ceramic bodies must comply all  
999 requirements, from milling to firing, to be transferable to tile manufacturing. In particular, rheological  
1000 behavior of slips and powder behavior during compaction frequently represent the bottleneck in the  
1001 scale up from lab to factory. Apparently unimportant, also the need of preliminary treatments is  
1002 fundamental to make the waste use viable, as it may determine a supply gap that prevents recycling.  
1003 It is not by chance that successful examples have generally no supply gap, so enabling a direct  
1004 delivery of residues to the tile-making plant. A simple preliminary treatment, e.g. drying or  
1005 comminution, constitutes a minor gap, if manageable by either the waste supplier or the end user.  
1006 However, when a selective removal of undesired components is necessary, beneficiation processes  
1007 must be set up with specific know-how and plants. This is typically a severe hindrance that requires  
1008 a synergic approach, for instance by involving raw materials suppliers (which can upgrade wastes  
1009 into industrial minerals) or collecting centres where residues can be transformed in secondary raw  
1010 materials suitable for ceramic tiles.

1011 The integration of the available literature data with the known industrial achievements allowed  
1012 attributing a Technology Readiness Level to recycling, waste by waste, with caution depending on  
1013 production technology and type of tile. The higher TRLs, relative to the industrial practice, are related  
1014 to wastes with composition and technological behavior close to those of ceramic raw materials, such  
1015 as granite recovered from quarry dumps; glass cullet from sorting of municipal solid wastes; fired  
1016 and unfired ceramic residues. An intermediate level is represented by wastes suitable for recycling  
1017 but not yet entered into industrial production, mostly because of technological obstacles, such as:  
1018 supply gap, difficulties in batch design due to the exotic composition (e.g., high Ba and Sr of screen  
1019 glass) or release of harmful substances during firing. Other residues were tested in industrially  
1020 relevant environment (pilot or full scale) including ornamental stone cutting and sawing sludge, coal  
1021 fly ash, spent foundry sand, ceramic tile polishing sludge, asbestos-containing demolition wastes,  
1022 sewage sludge and cathode tube glass. The issues preventing their recycling span from the behavior  
1023 during the ceramic process and the need of preventive treatments to the occurrence of hazardous  
1024 elements and potentially pollutant emissions during firing.

1025 At the lowest TRLs, wastes were evaluated just at the laboratory scale. Examples encompass mining  
1026 tailings, oil refining sludge, titania sludge, coal bottom ash, biomass ash, metallurgical slag, galvanic  
1027 sludge, dredging soil. In these cases, the criticalities seem to concern more the waste pre-treatment  
1028 and environmental issues (handling problems for bad smell, fermentation, acid release or possible  
1029 leaching of hazardous substances or harmful gaseous emissions) than the ceramic manufacturing.  
1030 The recommended percentage of waste recyclable is usually overestimated at low TRL. This  
1031 prompts an extreme caution in extrapolating technological feasibility on the basis of results obtained  
1032 at the laboratory scale only.

1033 From a circular economy perspective, it can be concluded that the use of waste in ceramic tiles is  
1034 technologically feasible and already in place for those residues that exhibit high affinity with the raw  
1035 materials they are replacing. This circumstance paves the way for making ceramic tile production,  
1036 which consumes raw materials for ~250 million tons per year worldwide, an important target of waste  
1037 recycling. On the other hand, much can still be done, by scientific and technological research, to  
1038 improve knowledge on residues tested at the lower TRLs.

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## *Supplementary material to Waste recycling in ceramic tiles: a technological outlook*

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### *Technological behavior of wastes in tile-making*

The ceramic industry has been recycling wastes, mostly represented by in-house residues coming from its own manufacturing cycles. This cannibalistic loop entails both solid and liquid residues: the former consist essentially of powders and semi-finished products, while the latter are basically wastewaters recovered after cleaning operations along the production lines. Scraps of fired tiles can be also recycled as a sort of moderately fluxing ingredient of vitrified tiles or filler in case of porous tiles. All these residues come back to the department of body preparation, where enter again the production flowchart. In addition, both pre-consumer (e.g., scraps and kiln furniture from sanitaryware plants) and post-consumer wastes (recycled glasses from urban or WEEE sorting) are frequently added to ceramic tile batches as well (Andreola et al., 2016; Silva et al., 2017). Different is the case of unsold finished products, which can be recycled (after appropriate grinding) in a range of applications in civil engineering (Silvestre et al., 2013; Medri and Landi, 2014; Penteado et al., 2016) including the addition in ceramic batches, as the above-mentioned fired scraps.

This overall approach is extensively pursued in Italy, where the average amounts of recycled materials are 11% (unfired) plus 2% (fired scraps) of the total body mass (Resca et al., 2015). The corresponding index of recycling is close to 100% for both solid and liquid residues (i.e., almost every waste is reused in the same production cycle). In practice, the only in-house waste material currently non-recyclable is the spent lime from scrubbers of kiln flue gases

(Dondi et al., 1990; Andreola et al., 1992a, 1992b, 1993; Rambaldi et al., 2016). Nevertheless, the implementation of such a recycling loop is viable only in presence of the entire manufacturing process: “partial cycle” plants (i.e., from press to kiln, fed with powders acquired from outside) must devolve their residues to “full cycle” plants having also the body preparation department (e.g., mill and spray drier). It is not a case that “full cycle” plants register a recycling index over 100% (on average 129%) because they are utilizing also wastes from other industries (Resca et al., 2015).

#### *A. Wastes from mining, quarrying and treatment of minerals*

There are many case-studies dealing with recycling in ceramic tiles of residues from mining, quarrying and treatment of minerals. Despite the good number of papers, the level of knowledge is essentially incremental and limited to laboratory scale testing. Waste recycling was mainly addressed to floor and wall tiles (mostly red firing) with a limited concern to porcelain stoneware bodies. However, several papers adopted firing conditions incompatible with current technologies in tile-making, turning results hard to be considered.

Residues stemming from mining and quarrying activities can be distinguished upon the stage in which they are produced: (a) during excavation; (b) after washing and cleaning of minerals; (c) as tailings of beneficiation processes; (d) after cutting and sawing of ornamental stones. However, despite these operations are industrially well established, quarrying and

mining residues offer an overall picture characterized by a strong compositional heterogeneity, which reflects the variety of parent rocks. For a detailed discussion on the different chemical compositions, see the text.

*A1. Wastes generated during excavation* (mostly discarded blocks and rock dust) are generally utilized on site for fillings and environmental remediation. Two exceptions concern ceramic raw materials: blocks of ornamental stone (granite) accumulated around quarries and marble rock dust. The best example of successful valorization of the waste granite blocks is that at the Montorfano and Monte Camoscio quarries, northern Italy (Dino et al., 2012). Here, granite blocks are recovered from quarry dumps and undergo classical mineralurgical treatments for feldspathic raw materials, entailing grinding, sieving, milling, magnetic separation, flotation. The products so obtained have been commercialized in the last two decades as quartz-feldspathic materials and quintessentially behave as flux in bodies for vitrified tiles (Carbonchi et al., 2003). This was confirmed by further case-studies on granite (Ghiani et al., 1997; Hernandez-Crespo et al., 2001; Echeverrigaray et al., 2016) or alkalisyenite mining residues (Il'ina and Lebedeva, 2010) tested as flux substitutes in ceramic batches (10–35%). Suitable properties were found for both porous wall tiles (Il'ina and Lebedeva, 2010; Echeverrigaray et al., 2016) and vitrified floor tiles (Ghiani et al., 1997; Hernandez-Crespo et al., 2001) with a recommended waste amount in the 10–20% range. Carbonate-rich residues were proposed as pore-forming additive in porous tiles in substitution of calcite or dolomite: marble rock dust (Montero et al., 2009a); dolostone (Amin et al., 2011) and magnesite (Binal and Ay, 2014). This solution can find practical application, with similar outcome, when carbonate-rich clays are not available.

*A2. Mining and quarrying wastes from washing, cleaning, and de-dusting* operations are essentially aimed at removal of the finest particle fractions and/or settling of the coarsest grain

fractions. In general, the expected role in ceramic batches is either as plasticity-provider (fines) or filler (sands) or pore-forming (carbonates). Clay materials were recovered from sand washing (Dondi et al., 1992; Danasino and Di Primio 1995; Sokolar and Grygarova, 2013; Sanchez-Soto et al., 2018) and after kaolin (Menezes et al., 2005, 2008; Varela et al., 2009), slate (Campos et al., 2004), oil shale (Melnick et al., 2010), serpentinite (Diaz and Torrecillas, 2007), or gold (Özkan et al., 2010) mining operations. Despite the mineralogical composition is comparable to clay raw materials, there may be limitations due to the high content in iron oxide (Melnick et al., 2010; Özkan et al., 2010; Sokolar and Grygarova, 2013) or muscovite (Menezes et al., 2008; Varela et al., 2009).

Iron-rich clay wastes were addressed to split tiles (Sokolar and Grygarova, 2013) and red stoneware (Melnick et al., 2010) with good technological outcome or, in alternative, as small addition (<10%) in white-firing bodies (Özkan et al., 2010). Kaolin waste was tested in both porous (Menezes et al., 2008) and vitrified tiles (Menezes et al., 2005; Varela et al., 2009) with a recommended amount from 25% to 60%. However, the well-known problem in managing muscovite in the tile-making process was not raised. An additional issue, not considered in the literature, is the content of water and organic matter of these sludges, which can be rather high but must be reduced to the level usually tolerated in ceramic tile manufacturing.

Agate rejects were tested in substitution of quartz sand (up to 65%) in porcelain-like batches by the design of experiments method (Correia et al., 2009). Satisfactory results were found with 15–22% of agate scraps.

*A3. Wastes from mineral beneficiation*, which commonly involves a range of physico-chemical processes (comminution and sieving, magnetic and electrostatic separation, flotation, acid leaching, and so on), are frequently referred to as *tailings*. These wastes are usually fine-grained

and characterized by the composition of gangue minerals plus possible contaminants deriving from mineralurgical treatments. This singles out issues that may hinder the utilization of tailings in tile-making: water content, pH, hazardous components (from ore and/or additives used in the various processing steps) and overall a composition that can be far from that of ordinary ceramic raw materials. To this regard, a distinction is necessary between two kinds of tailings from ore beneficiation and borate processing.

*Tailings from ore beneficiation* include residues from treatment of iron ore (Chen et al., 2013; Das et al., 2000, 2012; Fontes et al., 2019), Au-Cu-Mo ore (Mehrabian and Sarrafi, 2015; Solismaa et al., 2018; Karhu et al., 2019) and coal (Wei et al., 2010; Meseguer et al., 2009). In addition, residues from beneficiation of industrial minerals, such as apatite, bauxite, mica, quartz, feldspar, wollastonite (Kaya et al., 2004; Yang et al., 2009; Solismaa et al., 2018; Karhu et al., 2019; Hossain et al., 2019) and gemstones (Cavalcante et al., 2012; Pashkevitch

and Petrova, 2019). Beyond the chemical and mineralogical variability from case to case, overall traits are fine particle size, high amount of  $\text{Fe}_2\text{O}_3$ ,  $\text{MgO}$  and  $\text{CaO}$ , apart feldspar (Kaya et al., 2004; Karhu et al., 2019) and coal tailings (Wei et al., 2010; Meseguer et al., 2009). This circumstance restricts to applications in red-firing bodies, where tailings were tested as batch ingredient up to 70%. The fine particle size brings about some advantages during processing (better grindability, adequate plasticity and dry strength) and a generic role of fluxing agent, even if variation of firing behavior is often modest (Fig. A). However, the tiles containing iron ore tailings are easily exposed to overfiring, due to a narrow sintering interval and drastic fall of the physical and mechanical properties if temperature slightly overpasses the optimal range (Fig. A). Recommended percentages are in the 40–65% range for iron ore tailings (Chen et al., 2013; Das et al., 2000, 2012; Fontes et al., 2019) and 10–34% in case of coal, wollastonite and emerald tailings (Wei et al., 2010; Cavalcante et al., 2012; Hossain et al., 2019).

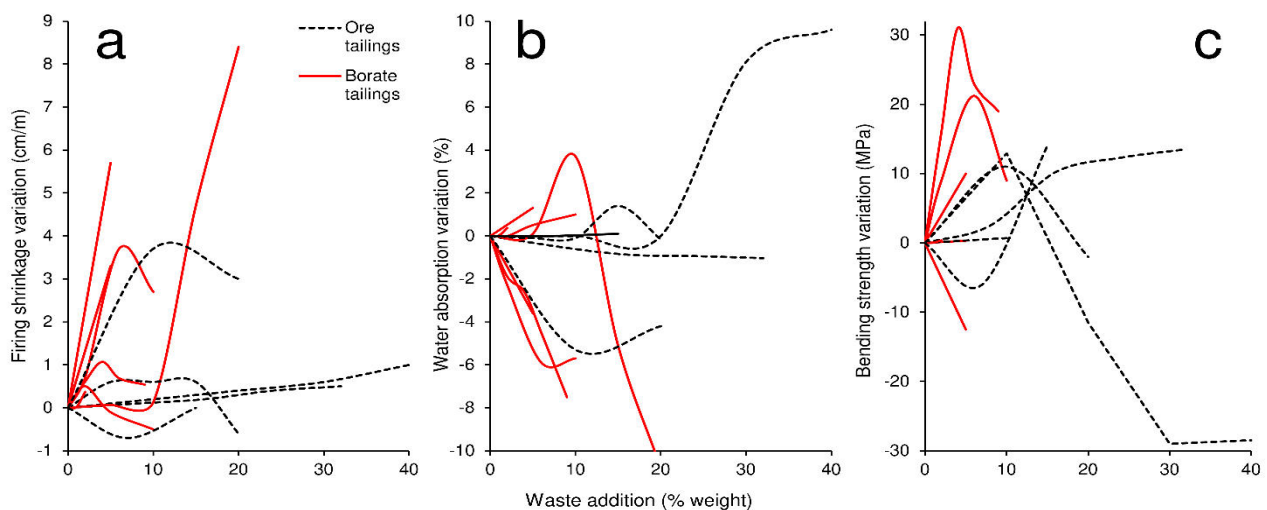


Figure A. Effect of mining tailings on the firing behavior of ceramic tile bodies. Variation of: a) linear shrinkage, b) water absorption, c) bending strength, as a function of waste amount. Borate tailings (Olgun et al, 2005; Kurama et al., 2007; Ediz and Yurdakul, 2009; Celik, 2015; Özkan, 2017; Zanelli et al., 2019); other ore tailings (Kaya et al., 2004; Özkan et al., 2010; Cavalcante et al., 2012; Das et al., 2012; Chen et al., 2013).

*Tailings from borate processing* are always characterized by an important concentration of  $\text{B}_2\text{O}_3$  (4–32%) that strongly affects their technological behavior (Olgun et al, 2005;

Kurama et al., 2007; Ediz and Yurdakul, 2009; Cetin et al., 2015; Celik, 2015; Özkan, 2017; Tang et al., 2018; Korç et al., 2018; Cicek et al., 2018; Karadağlı and Cicek, 2019; Zanelli et al.,

2019). Such residues are featured by borates (tincalconite, ulexite, colemanite) and a high amount of alkali-earths – due to magnesite, calcite, plagioclase, chlorite – along with clay minerals and quartz. Sulfur may be present in high concentration, not compatible with the gaseous emission threshold from kilns.

These tailings were tested, generally in percentages below 20%, either in wall tiles (Olgun et al, 2005; Kurama et al., 2007; Celik et al., 2014; Cetin et al., 2015; Celik, 2015; Özkan, 2017; Cicek et al., 2018) or in porcelain stoneware (Ediz and Yurdakul, 2009; Korç et al., 2018; Karadağlı and Cicek, 2019; Zanelli et al., 2019). There is an overall convergence to a major fluxing effect, with increased reactivity during firing and possible reduction of the maximum temperature (Fig. A). However, changes in the milling and compaction behavior are limited. Batches containing tailings up to 5% (porcelain stoneware) or 10% (porous tiles) have a suitable technical performance, even though these sludges tend to lower bulk density and increase closed porosity (Zanelli et al., 2019).

*A4. Sludges from ornamental stone cutting and sawing operations give rise to fine-grained sludges with a composition close to that of*

ornamental stones, but containing processing residues, like scraps of grinding media and additives (flocculant, etc). This justifies higher percentages in Fe<sub>2</sub>O<sub>3</sub> and CaO, and raises issues habitually related to the occurrence of too high values of pH and/or iron and calcium oxides (boosting slip viscosity, dark firing color). The compositional range is rather wide, comprising granite (Torres et al., 2004, 2007; Menezes et al., 2005; Vieira and Monteiro, 2006; Souza et al., 2008; Hojamberdiev et al., 2011; Silva et al., 2011; de Almeida et al., 2012; Gadioli et al., 2012; Aguiar et al., 2014), gneiss (Souza et al., 2010a, 2010b, 2011; Junkes et al., 2011, 2012), quartzite (Souza and Costa, 2015; Biff and Silva, 2016; Medeiros et al., 2017) on one side and limestone-marble-doloma on the other side (Montero et al., 2009a; Jordan et al., 2015). Thus, the expected technological role goes from flux to filler, depending on the feldspars-to-quartz ratio, or even pore-forming agent in case of carbonate-rich residues. Waste additions were experimented up to 75% of the batch in both vitrified (Hojamberdiev et al., 2011; Silva et al., 2011; Junkes et al., 2011, 2012) and porous tiles (Menezes et al., 2005; Montero et al., 2009a; Gadioli et al., 2012; Jordan et al., 2015; Marvila et al., 2018).

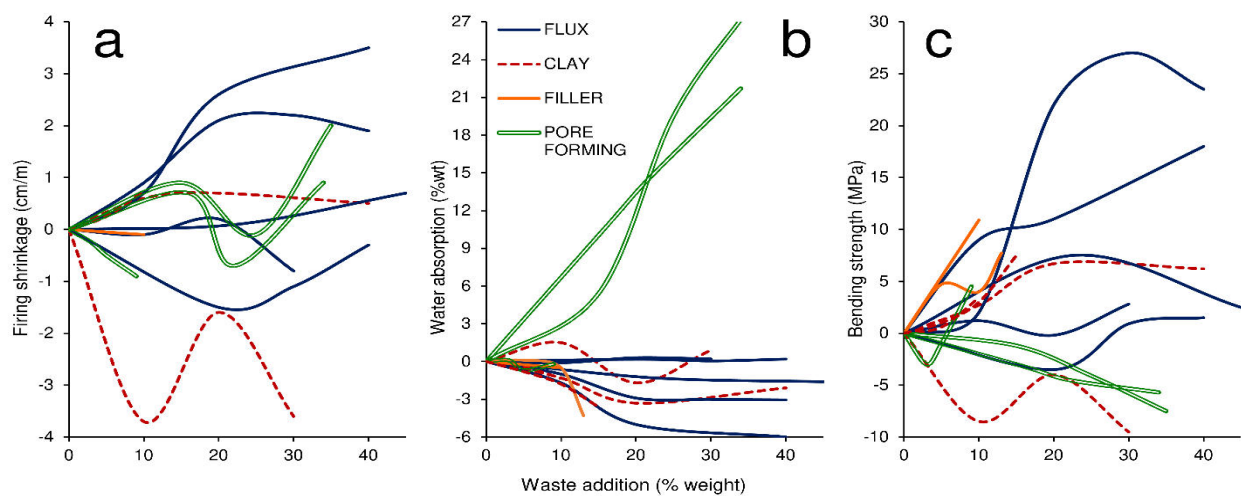


Figure B. Effect of ornamental stone cutting and sawing sludge on the firing behavior of ceramic tile bodies. Variation of: a) linear shrinkage, b) water absorption, c) bending strength, as a function of waste amount. Waste addition in replacement of flux (Torres et al., 2004; Vieira and Monteiro, 2006; Souza et al., 2008, 2010a; Silva et al., 2011), filler (Menezes et al., 2005; Hojamberdiev et al., 2011), clay (Gadioli et al., 2012; Echeverrigaray et al., 2016; Biff and Silva, 2016; Medeiros et al., 2017), or pore-forming agent (Montero et al., 2009a; Amin et al., 2011; Jordan et al., 2015).

In these papers, technological results are apparently contradictory, as they reflect the variability of lithologies and testing conditions, especially batch design and firing cycles (Fig. B). Various strategies were followed in batch design: adding the sludge in substitution of either a clay (in a binary mix) or flux or filler (in a complex formulation). When a granite-gneiss sawing sludge is added to a clay body, an expected reduction of plasticity (Menezes et al., 2005; Junkes et al., 2012; Aguiar et al., 2014; Medeiros et al., 2017) and drying shrinkage (Torres et al., 2004; Silva et al., 2011) is recorded, together with a decrease in water absorption associated to an increase of firing shrinkage and bulk density (Menezes et al., 2005; Torres et al., 2007; de Almeida et al., 2012; Gadioli et al., 2012; Medeiros et al., 2017) while bending strength can either increase or decrease (Fig. B). In complex batches, where the sludge replaced a flux, a drop in dry bulk density was observed until 10–20% waste, even if values increase up to 40% (Torres et al., 2004; Vieira and Monteiro, 2006; Souza et al., 2010b; Silva et al., 2011). After firing, waste-bearing bodies exhibit a lower or similar water absorption (Fig. B) and values of shrinkage and bending strength higher or analogous to the waste-free batches (Torres et al., 2004; Menezes et al., 2005; Vieira and Monteiro, 2006; Souza et al., 2010a; Silva et al., 2011). The recommended percentages are in the 15–70% range for granite sludge in replacement of clay (Torres et al., 2007; Menezes et al., 2008; Gadioli et al., 2012; de Almeida et al., 2012; Medeiros et al., 2017) but generally below 10% once a granite sludge (Souza et al., 2008; Silva et al., 2011) or a sand and quartzite residue (Souza and Costa, 2015; Biff and Silva, 2016; Medeiros et al., 2017) is substituted after feldspar.

### ***B. Wastes from the pulp and paper industry***

The residues from the cellulose and paper production that have been investigated as ceramic raw materials consist in paper mill

sludge (Asquini et al., 2008; Maschio et al., 2009; Furlani et al., 2011; Sahar et al., 2011), cellulose sludge (Azevedo et al., 2019), and grits (Siqueira and Holanda 2018). The mill sludge cannot be recycled as a sludge, and must be first treated (de-watering, incineration, and milling). Account must be taken that heavy metals (Ba, Cr, Cu, Pb, Sn, Sr, V) can be found in significant amount in the ash. This residue was tested at the laboratory scale, alone or mixed with soda-lime glass and clay (10–60%). An increase in water absorption and firing shrinkage can be attributed to the paper waste, while mechanical behavior did not greatly differ (Maschio et al., 2009; Furlani et al., 2011). Since data on ceramic tile batches containing only paper ash are missing, results are inconclusive about the technological feasibility.

Other wastes from the pulp and paper industry consist largely of calcium carbonate. Grits is a noninert solid waste derived from the liquor causticizing step in cellulose production. It was tested up to 15 wt% in replacement of calcium carbonate used as pore-forming agent in ceramic wall tiles (Siqueira and Holanda 2018). The results revealed that despite this waste promotes the formation of porosity, leading to lesser dense bodies, the bending strength is enhanced. This occurs probably in force of a microstructural improvement and formation of anorthite and mullite during fast firing at 1100–1180°C.

Another study incorporated the paper residue up to 30% into a batch for split tiles (Azevedo et al., 2019). Waste-bearing bodies exhibit higher water absorption and lower firing shrinkage, as expected, but a lower total porosity and better bending strength for the addition of 10% residue. Hence, this kind of paper wastes could be used as a total replacement of calcareous raw materials traditionally added to wall tile batches, when carbonatic clays are not available.

### **C. Wastes from petroleum and edible oil refining**

Oily sludge, a hazardous material produced during oil extraction, was extensively investigated (Pinheiro and Holanda, 2008; Pinheiro et al., 2013; Pinheiro and Holanda, 2013a, 2013b). It is basically a drilling mud contaminated by petroleum. Thus, the sludge is based on bentonite, barite (BaO at 5%) plus additives and fragments of drilled rocks, which all contribute to the high loss on ignition. Tests at the laboratory scale were conducted adding the sludge to a porcelain stoneware batch in replacement of kaolin up to 5-10% (Pinheiro and Holanda, 2008; Pinheiro et al., 2013; Pinheiro and Holanda, 2013a, 2013b). Dry bending strength slightly increased with the sludge addition, as expectable by its bentonite content, but no significant difference was observed in drying shrinkage and dry bulk density of tiles. Fast firing revealed a decremental effect on technological properties as a function of sludge percentage: bulk density and bending strength decreased, while water absorption increased. A rapidly decreasing firing shrinkage likely denotes a tendency to bloat, justified by the content in  $\text{Fe}_2\text{O}_3$  and organic matter. Nevertheless, such changes are moderate for small amounts of waste: an oily sludge addition up to 2.5% is considered tolerable for porcelain stoneware tiles (Pinheiro and Holanda, 2008; Pinheiro et al., 2013; Pinheiro and Holanda, 2013a, 2013b). Leaching tests gave values of hazardous elements within regulatory limits (Pinheiro and Holanda, 2013a).

Another oily sludge, recovered after processing of palm oil, is expected to be a nonhazardous material with a composition compatible with silicate ceramics. It was introduced up to 20% in a porcelain stoneware for artware (Salehi et al., 2015) and tested at the lab scale, even in combination with a waste glass (Salehi et al., 2016). A conclusive word on technological feasibility is not possible, because little is known about the behavior during processing and tiles

were fired with a slow cycle. Anyway, water absorption decreased, and firing shrinkage increased, as a function of sludge amount, up to 10%. Beyond this percentage, there are clues of overfiring (Salehi et al., 2015).

### **D. Wastes from inorganic chemical processes**

In this category are comprised residues stemming from industrial processes used to produce alumina, titania, zinc, phosphoric acid, or sodium silicate. Overall, they consist of “muds” equaled by a fine particle size distribution and relatively high moisture (semi-solid sludges). All the investigations were carried out at the laboratory scale.

Red mud is the main by-product of bauxite processing to get alumina by the Bayer cycle (Pontikes et al., 2007a; Yang et al., 2009; Wang et al., 2018). The major component is iron oxide together with alumina, CaO and silica. This fact restricts any application to red-firing products. The sintering behavior of red mud was tested up to 1100°C under different atmospheres (Pontikes et al., 2007a, 2009) also mixed with kaolin (Wang et al., 2018). Fired bodies are characterized by a compact heterogeneous matrix, with isolated closed pores. Phase composition has magnetite, wustite, gehlenite and perovskite as main crystalline phases, along with a low viscosity liquid phase form at high temperature (Pontikes et al., 2007a, 2009). Such microstructure results in improved physical properties (Pontikes et al., 2009). Another kind of red mud comes from hydrometallurgical processes of zinc ore (Kummoonin et al., 2014). It has a silicate basis, rich in Ca and Fe oxides with significant concentrations of heavy metals, like Zn, Pb, As, Cr, Cu, Ni and V (Hua et al., 2017; Khairul et al., 2019). As said about the Bayer red mud, the high amount of iron constrains the recycling to red-firing bodies or to tiny percentages in white-firing batches. It was tested (60-70%) in a vitrified tile batch – together with steel slag and recycled soda-lime glass – fast fired at 900-1050°C in a methane-fed furnace (Kummoonin et al., 2014). In front of a high



bulk density and 2.6% water absorption, firing shrinkage was very high (11%) and flexural strength quite low (26 MPa). Although encouraging, available data does not allow a conclusive deduction about the technological feasibility of red mud recycling.

A brown sludge (“tionite”) consists of unreacted residues after the sulfuric dissolution of ilmenite-titania slag to produce titania (Dondi et al., 2010). The main issues are the fair amount of SO<sub>3</sub> and a certain level of radioactivity (Contreras et al., 2014). Tionite was admixed up to 50% to an industrial red stoneware batch and fast-fired. A small addition (3-5%) had beneficial effects, easing both the drying and firing processes: water absorption was reduced, while bending strength improved (Contreras et al., 2014, 2016). The leaching tests demonstrated that the mobility of heavy metals and radionuclides is similar to that of the waste-free tile (Contreras et al., 2014). However, unpublished lab tests, adding tionite in porcelain stoneware, were unsuccessful because of intolerable effects in wet ball milling (boosting slip viscosity and thixotropy) even with few percent of waste.

Phosphogypsum is the main waste of the production of phosphoric acid (Contreras et al., 2018; Huang et al., 2020). It is basically composed of calcium sulfate, which is not compatible with the silicate batches used in tile-making, essentially because it starts decomposing already at 900°C (Coronado et al., 2016) with release of sulfur oxides in the kiln flue gases. In top of this, a safety issue is represented by the known radioactivity of phosphogypsum (Huang et al., 2020). It was added up to 10% to a red clay and slow fired at 950-1150°C. Waste addition promoted a better sintering: higher shrinkage, lower porosity and water absorption, along with no significant variation in the mechanical resistance (Contreras et al., 2018). The recommended amount (7.5% phosphogypsum) for recycling, even if technologically feasible in red stoneware, however, implies a noteworthy

hazard, as the potential release of SO<sub>x</sub> in the flue gases is one order of magnitude higher than the permissible emissions.

A sludge stemming from the production of sodium silicate (waterglass) was added (up to 20%) in a batch for floor tiles (Osman et al., 2008). Despite the expected fluxing role – based on high Na<sub>2</sub>O and CaO amounts and low percentage of alumina in the waste – resulting tiles exhibit increased water absorption combined with reduced bulk density and compressive strength. It is concluded that no more than 5% of sludge could be used to keep any change in technological properties tolerable.

Further experiments regarded an adsorbent from solvent cleaning and a residue of ferrite processing were tested as opacifiers in porcelain stoneware (Mestre et al., 2010).

#### ***E. Wastes from thermal processes***

Thermal power plants produce energy by burning organic matters of various types, from naturally occurring coal to sewage sludge, municipal solid wastes, and biomasses resulting from the processing of a multitude of starting materials. Combustion processes give rise to unburnt residues that can be basically distinguished, on the basis of their granulometry, between coarse-grained bottom ash and grate slag (e.g., Barbieri et al., 2002; Bourtsalas et al., 2015a) and fly ash that generally has a very fine particle size (e.g., Dana et al., 2005; Wang et al., 2017). The main chemical traits are inherited from the thermal plant raw materials (coal or other fuels). Despite the rather large number of papers, the knowledge about the ash effect in ceramic tile bodies is essentially limited to the laboratory scale, with few examples at a pilot plant (e.g., Mishulovich and Evanko, 2003; Ji et al., 2016; Luo et al., 2017a). Coal ashes have been extensively investigated, pursuing various strategies. Being composed by about 90% of silica, alumina, CaO and Fe<sub>2</sub>O<sub>3</sub>, and relatively poor in alkalis, coal ashes are compatible with ceramic batches, particularly those for porous

bodies. A matter of concern is the amount of some heavy metals (As, Cr, Cu, Ni, Zn) particularly in fly ash (Pandey et al., 2011; Tsiridis et al., 2012). Since the iron content is rather high, the subsequent coloration of the ceramic body is generally dark. Therefore, the target was mainly constituted by bodies for red stoneware, together with porous red tiles or simple red clay:ash mixes for split tiles. Attempts to recycle coal ash in porcelain stoneware are rare. A case apart are papers dealing with waste-based bodies.

*E1. Coal bottom ash* was investigated as an ingredient in different ceramic tile batches: in substitution of kaolin, pottery stone, feldspar and quartz (up to 50%) in light-firing stoneware (Prasartseere et al., 2018); in ash:clay (up to 80%) and ash:clay:sand mixes (up to 40%) for red stoneware (Namkane et al., 2016, 2017); added (up to 30%) to a porous red body (Skoronski et al., 2015) even together with a borate tailing (Bayca et al., 2008). No information is available on the pre-heating behavior, apart the observation that bottom ash enhanced the time-of-flow of slips, likely in force of an increased viscosity (Skoronski et al., 2015). The effect of bottom ash during firing cannot be generalized: firing shrinkage, water absorption and bending strength were found to vary upon the waste source and the tile typology. For instance, lignite bottom ash, substituting kaolin and/or pottery stone, promoted a lower pyroplastic deformation, even though some batches denoted a water absorption higher than the standard stoneware tile. The more the lignite bottom ash, the more the anorthite phase and the less the pyroplastic deformation (Prasartseere et al., 2018). A relevant problem may be the black core, due to the occurrence of unburnt carbon residues in the ash (Skoronski et al., 2015). Another case is about ash admixed with other wastes in order to exploit a synergistic effect. Adding 15% of bottom ash together with up to 3% of borate tailing, for example, had a favourable effect on the physical properties of wall tile bodies. Both

water absorption and firing shrinkage decreased with increasing waste content (Bayca et al., 2008). Overall, the recycling of coal bottom ash was considered technologically feasible, with a recommended amount within 10% in light-firing stoneware (Prasartseere et al., 2018); 10–15% in wall tiles (Bayca et al., 2008; Skoronski et al., 2015); 20–33% in red stoneware (Namkane et al., 2016).

*E2. Coal fly ash* attracted a wide research interest. It was tested in replacement of feldspar and quartz (up to 20%) in porcelain stoneware (Dana et al., 2005; Kockal, 2012); in substitution (up to 40%) of clay (Rajamannan et al., 2013; Kim et al., 2017) or up to 10% instead of feldspar (Olgun et al., 2005) in porous tiles; as clay-ash mixes (up to 90%) in stoneware-like or split tiles (Mishulovich and Evanko, 2003; Zimmer and Bergmann, 2007; Yürüyen and Toplan, 2009; Sokolar and Smetanová, 2010; Sokolar & Vodová, 2011; Trnik et al., 2013; Michalíková et al., 2014; Ji et al., 2016; Wang et al., 2017). In addition, special batches were experimented, based on fly ash only (Luo et al., 2017a, 2018) or admixed with other residues (Sokolar, 2007; Tan and Wang, 2011) or talc (Chandra et al., 2005). Fast firing was applied only in two cases (Mishulovich and Evanko, 2003; Kockal, 2012). The effect of fly ash pre-treatment was seldom considered, focusing on milling and alkali activation (Luo et al., 2017a, 2017b). However, milling is generally mandatory to improve sintering, and a residue of maximum 5% on a 63 $\mu$ m sieve is recommended (Sokolar, 2007; Sokolar and Smetanová, 2010). On the other hand, fly ash seems to improve grindability in simple waste:clay mixtures (Zimmer and Bergmann, 2007). Information on powder behavior during compaction is lacking, while that on drying is limited to a single observation, where a small fly ash addition in replacement of feldspar did not change the drying shrinkage (Olgun et al., 2005). As observed for bottom ash, the effect of fly ash on firing cannot be stereotyped:

technological parameters exhibit contrasting trends that seem to depend on the kind of ceramic tile and/or the raw material replaced (Fig. C). In any case, the specific features of each fly ash are crucial, as water absorption or

bending strength were found to increase or decrease as a function of waste addition even in the same batch type: e.g., porcelain stoneware (Dana et al., 2005; Kockal, 2012) or porous tiles (Olgun et al., 2005; Kim et al., 2017).

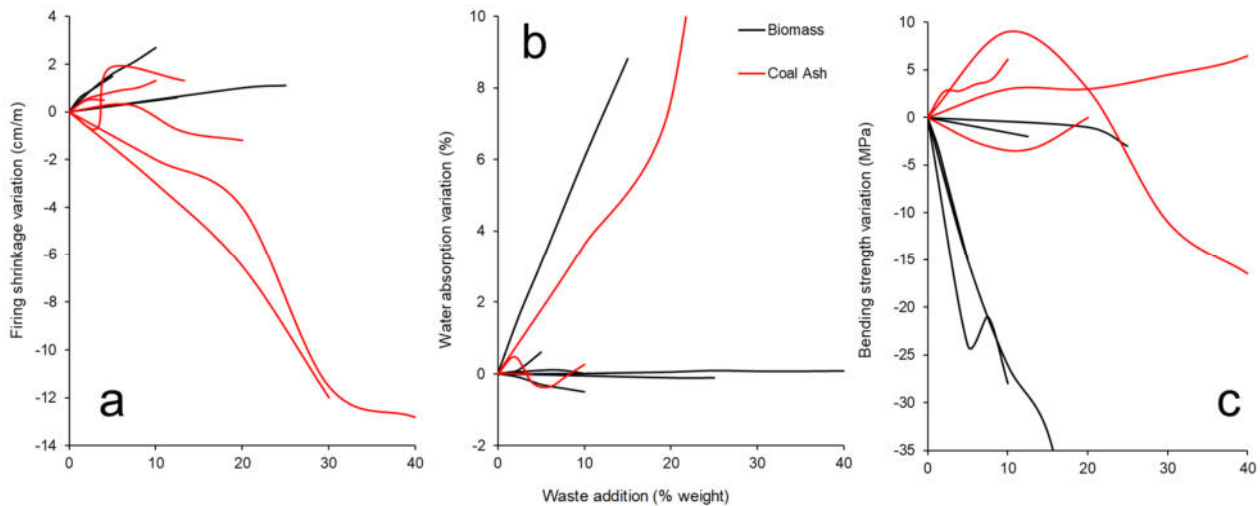


Figure C. Effect of coal or biomass ashes on the firing behavior of ceramic tile bodies. Variation of: a) linear shrinkage, b) water absorption, c) bending strength, as a function of waste amount. Biomass (Amutha et al., 2014; Guzman et al., 2015; Guzman et al. 2016; Naga and El-Maghraby, 2002; Novais et al., 2015; Olokode et al., 2013; Paranhos et al., 2017; Schettino and Holanda, 2015), Coal ash (Boutsalas et al., 2015a; Dana et al., 2005; Ji et al., 2016; Namkane et al., 2016; Namkane et al., 2017; Olgun et al., 2005; Wang et al., 2017).

A particular concern must be devoted to the risk of black core formation and bloating, especially in case of fast heating and cooling rates (Sokolar, 2007; Sokolar and Smetanová, 2010). This risk is connected with the high content of iron oxide of fly ash and notably of carbon residues, which can attain 14–19% (Kim et al., 2017). Fly ash can play as sintering promoter in porcelain stoneware, but it seems in relation to the rather high alkaline–earth oxide content, which favors an early maturing of bodies by their strong fluxing action (Dana et al., 2005). Nevertheless, replacing feldspar with 10% fly ash induced acceptable changes in technological properties, even though porosity increased (Kockal, 2011). In wall tiles, the addition of fly ash had limited effects: water absorption slightly changed, with increasing waste content up to 30%, while bending strength of the tile including 10% fly ash was at a level comparable to the ash-free batch (Kim et al., 2017). Amongst the experiments mixing clay and fly ash, two were

run at the pilot scale, using commercial equipment for mass production and white-firing ball clays (Mishulovich and Evanko, 2003; Ji et al., 2016). A technological feasibility was demonstrated, including wet ball milling and fast firing, for a concentration of fly ash up to 40–65%. Tests carried out with waste-based batches gave rather optimistic assessments on their industrial transferability, stating the possibility of using 100% fly ash with a trial on a pilot plant (Luo et al., 2017a). Another case is fly ash mixed with talc: on its gradual addition, an improvement in the impact strength was observed, while water absorption decreased to a minimum for a waste:talc ratio of 40:60% (Chandra et al., 2005).

In conclusion, although there is a general propensity to consider the recycling of fly ash (and bottom ash) as technologically feasible, tests conducted in the conditions of modern tile manufacturing are almost completely lacking. It is probably the difficult management of

particularly fine and volatile powders, as well as the presumable presence of carbonaceous unburnt materials, which ensures that there are no examples, to the best of our knowledge, of industrial production with the use of fly ash, although this recycling is in use in the brick industry for at least 30 years (Dondi et al., 1997).

*E3. Sewage sludge ash* chemical features are strongly variable according to the sources. Sewage ash was experimented following various strategies: alone (Lin et al., 2006); added with nanosilica (Chen and Lin, 2009; Lin et al., 2016); or admixed in high percentage (minimum 25%) to clay materials (Lin et al., 2008) and further wastes, like soda-lime glass (Merino et al., 2007) or steel slag (Favoni et al., 2005). No investigation was carried out in industrial tile-making conditions. Preliminary treatments consisted essentially in drying and grinding the agglomerated ash, e.g. by ball milling and sieved <math><150\mu\text{m}</math>. Vitrified bodies (bulk density 2.28–2.36 g·cm<sup>-3</sup>) were obtained after firing the ash at 1000°C (Lin et al., 2006) or 1200°C (Merino et al., 2007). However, no generalized technological behavior can be inferred, because apparently contrasting results were achieved, depending on the clay types and waste additions. For instance, the best compressive strength was obtained with mixes 75% ash + 25% clay (montmorillonite or illite, but not kaolin, due to its refractoriness) possibly with powdered flat glass (Merino et al., 2007). Interestingly, nano-SiO<sub>2</sub> addition had a positive influence on the properties of mixes where potter's clay or porcelain clay were replaced with incinerated sewage sludge ash. Nevertheless, care should be taken to optimize both clay-ash ratio and firing temperature (Chen et al., 2009), as the optimum kiln temperature varied between 1100 and 1150°C (Lin et al., 2016). Moreover, both water absorption and bending strength were affected by glazing (Lin et al., 2008). Overall, it is not possible to solve the doubt about the feasibility of recycling ash from sewage sludge in ceramic tile production, although there are promising indications from

the literature tests. In fact, too many aspects have not been evaluated, starting from possible interferences in the grinding and pressing processes, up to the role of phosphorus and iron during firing, and the effective stabilization of heavy metals.

*E4. Ashes from municipal solid waste incinerator* (MSWI) are special wastes to be managed in form of fly ash or bottom/grate ash. Fly ash was utilized (up to 30%) as clay substitute in porous tile (Haiying et al., 2007; Jordán et al., 2015) or porcelain stoneware bodies (Hernández-Crespo and Rincón, 2001; Hernández-Crespo et al., 2002). Bottom ash was added up to 20% directly to a porcelain stoneware batch (Andreola et al., 2001, 2002). In alternative, previously vitrified bottom and grate ashes were tested in porcelain stoneware, as a replacement (up to 10%) of feldspar (Rambaldi et al., 2010) or added up to 10% to the body (Barbieri et al., 2002) or to a clay (Schabbach et al., 2012). The same vitrified bottom ash was also experimented (5%) in a porous tile body (Rambaldi et al., 2010). Finally, MSWI bottom ash was investigated mixed with up to 30% soda-lime glass (Bourtsalas et al., 2015a) or added to the sludge stemming from porcelain stoneware tile polishing in the 10:90 ratio, with an extra 10% to 20% silica addition, to produce lightweight vitrified ceramic tiles (Tan and Wang, 2011). MSWI ashes exhibit chemical features that heavily constrain their utilization as ceramic raw materials. Apart from the abnormally low values of silica and alumina, the concentration in iron oxide is a strong limit to the use in light-firing bodies. More important is to keep under control the modest concentration of heavy metals (Ba, Cd, Cr, Cu, Hg, Mn, Pb, Sn, Zn) and, above all, the remarkable quantities of chlorine and sulfur (Zacco et al., 2014; Zhou et al., 2015). These components can turn intolerable in relation to the release by leaching and the gaseous emissions during firing, respectively. MSWI bottom ashes are typically finer than 4 mm and can be directly mixed with clay and non-plastic ingredients.

Otherwise, a preliminary treatment is needed, e.g., milling and calcination, in order to avoid excessive shrinkage and firing deformations (Bourtsalas et al., 2015a, 2015b). The direct mixing of bottom ash in the porcelain stoneware body induced an increase of the slip viscosity as a function of the waste amount. However, batches containing up to 20% of bottom ash exhibit tolerable variations of mineralogical, thermal, and rheological properties (Andreola et al., 2001). A comparison between the untreated bottom ash (after deferrization) versus vitrified ash let to a reduction in the heavy metals release, even though the thermal treatment is not sufficient to classify the resulting material as inert. The vitrified ash, once added to porcelain stoneware in substitution of 5–10% feldspar, fostered the crystallization of plagioclase and reduced the amount of vitreous phase. The

densification process is enhanced by lowering water absorption and increasing firing shrinkage and bulk density (Fig. D). A reduction of about 20°C in the maximum firing temperature is achievable, besides closed porosity could increase (Rambaldi et al., 2010). More elaborate glass recipes – where bottom ash was 50:50 with soda-lime cullet (Barbieri et al., 2002) or 60:40 with refractory clay (Schabbach et al., 2012) – brought to characteristics of the final product slightly away from the reference body. However, some worsening in tile planarity and a significant modification of the color must be reported (Barbieri et al., 2002). The technological properties match the ISO standards of BIb and BIIa groups, i.e. water absorption between 2 and 5% (Schabbach et al., 2012).

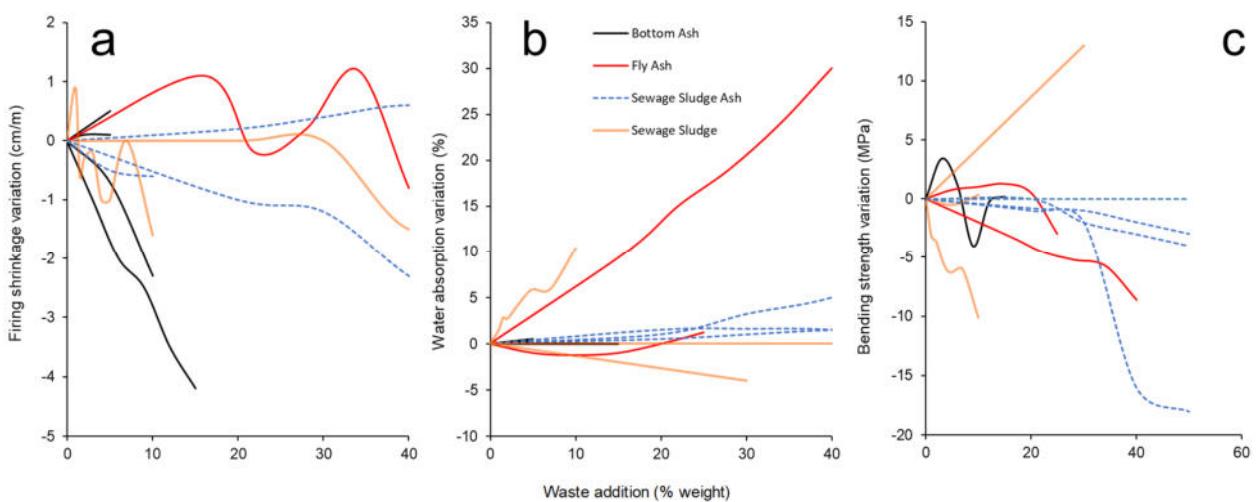


Figure D. Effect of MSWI bottom or fly ash or sewage sludge ashes on the firing behavior of ceramic tile bodies. Variation of: a) linear shrinkage, b) water absorption, c) bending strength, as a function of waste amount. Bottom ash (Barbieri et al., 2002; Rambaldi et al., 2010; Schabbach et al., 2012), Fly ash (Haiying et al., 2007; Jordán et al., 2015), Sewage sludge ash (Chen and Lin, 2009; Lin et al., 2008; Lin et al., 2016), Sewage sludge (Jordán et al., 2005; Zhou et al., 2013).

On the other hand, the addition of MSWI fly ash to porous tile batches promoted a decrease of water absorption and an increment of mechanical resistance, with acceptable results up to 20% at a sintering temperature of 980°C (Haiying et al., 2007). At all events, calcination generally improves the ash behavior, while recycling in ceramic bodies may bring about a

sort of encapsulation into glassy and/or crystalline phases, resulting in a minimal leaching compared to as-received bottom ashes (Bourtsalas et al., 2015a). As a matter of fact, the leaching of toxic elements from fly ash was significantly reduced through recycling in ceramic bodies, as the release of Cd, Hg, Pb and Zn turned within the permissible limits (Haiying

et al., 2007; Jordán et al., 2015). It can be concluded that recycling of MSWI bottom ash in ceramic tiles is technologically feasible, even better in case of upgrading to a vitrified byproduct. This is demonstrated also by the industrial scale-up of bottom ash to a commercial product (Matrix<sup>®</sup>) addressed, among other end-users, to tile manufacturers. In contrast, the picture about the MSWI fly ash is inconclusive since data available are far from current tile-making processes.

*E5. Combustion of biomasses* produces a wide range of ashes, investigated as possible raw materials for ceramic tiles. Sources are extremely varied, including the incineration of rice husk (Dana and Das, 2002; Andreola et al., 2013; Guzman et al., 2015, 2016; Abeid and Park, 2018), coffee husk (Acchar et al., 2013a; Acchar and Dultra, 2015; Acchar et al., 2016; Carvalho et al., 2017), sugar cane bagasse (Sivakumar et al., 2014; Schettino and Holanda, 2015; Faria and Holanda, 2016; Paranhos et al., 2017), palm oil and sago (Noh et al., 2014; Jamo et al., 2015; Aripin et al., 2010), cereal straw and stalk (Guzman et al., 2013; Amutha et al., 2014), wood and forestry wastes (Olokode et al., 2013; Novais et al., 2015), fish bone (Naga et al., 2014; Awaad et al., 2015) and leather waste (Fernandes and Ferreira, 2007). Burning biomasses gives rise to ashes with a considerably variable composition, primarily depending on the source. Some concern should be addressed to the amount of Cl and S (Niu et al., 2013) as well as to the fate of some heavy metals, like Co, Cr, Ni and Zn (Grammelis et al., 2006).

Among the undesired components, iron oxide is sometimes in significantly high amount for light-firing bodies. Also, sulfur and chlorine can be present in quantities that require careful evaluation of the risk of polluting emissions during firing. Moreover, biomass ashes may be particularly rich in organic residues, to the point to need a preliminary calcination before recycling (Noh et al., 2014).

Recycling of biomass ash was tried above all in porcelain stoneware bodies, where the waste

was introduced in substitution of feldspar up to 60% (Guzman et al., 2013, 2015), quartz up to 20% (Amutha et al., 2014; Schettino and Holanda, 2015; Abeid and Park, 2018), clay up to 40% (Olokode et al., 2013; Abeid and Park, 2018), phyllite up to 10% (Paranhos et al., 2017) or up to 25% as addition to the whole body (Fernandes and Ferreira, 2007; Sivakumar et al., 2014; Novais et al., 2015). Otherwise, these ashes were added (up to 50%) to clay-based stoneware (Aripin et al., 2010; Acchar et al., 2013; Naga et al., 2014; Awaad et al., 2015); up to 25% in replacement of quartz in porcelain batches (Noh et al., 2014; Jamo et al., 2015); up to 5% instead of clay in wall tiles (Faria and Holanda, 2016); as the only ingredient of glass-ceramic tiles (Andreola et al., 2013).

Substantially, no information was found about the technological behavior during body preparation, pressing and drying. However, several studies applied firing schedules comparable to current industrial practice (e.g., Schettino and Holanda, 2015; Guzman et al., 2015; Faria and Holanda, 2016; Paranhos et al., 2017; Abeid and Park, 2018). Although at a laboratory scale, biomass ashes seem generally adequate to replace fluxes or fillers in low proportion in glazed porcelain stoneware. A 50 to 100°C reduction of firing temperature is reported, without influencing too much the technical properties and the tile microstructure. This occurred with ashes from coffee husk, rice straw, and sugarcane bagasse, which are characterized by high alkali values (Acchar et al., 2013a; Acchar and Dultra, 2015; Schettino and Holanda, 2015; Guzman et al., 2015, 2016; Paranhos et al., 2017). Nevertheless, increasing the ash content above 25% significantly affected the properties of fired tiles, as result of the higher content of gas-generating substances (Guzman et al., 2013, 2016; Abeid and Park, 2018).

Positive results were registered also with other wastes, like palm oil ash, as a substitute of quartz in a porcelain batch, which led to an increase in compressive strength (Jamo et al., 2015). Also, sago ash in mixture with clay improved water

absorption and bulk density in floor tiles (Aripin et al., 2010).

Another ash obtained after combustion of leather residues, derived from the shoes industry, was tested up to 10% into a porcelain stoneware batch (Fernandes and Ferreira, 2007). No conclusion about technological feasibility is possible since no information is available about the behavior during processing and firing was carried out with a slow cycle. Nevertheless, there are evidences this waste plays as a strong flux, reducing by  $\sim 60^{\circ}\text{C}$  the temperature of maximum densification, in accord with a composition rich in Fe, Cr, Na and poor in Si and Al. A darker color after firing and a fast decrease in bulk density were observed for increasing ash amount (no more than 1% tolerable).

Although coffee grounds are not ashes, strictly speaking, they are here treated together with other biomass derived wastes. Grounds previously washed, dried, and crushed below  $63\mu\text{m}$ , were used up to 30% to produce wall tiles by firing at  $1150^{\circ}\text{C}$ . The coffee waste strongly increased porosity and water absorption while decreased bulk density and flexural strength (Manni et al., 2019). Acceptable technological properties (especially flexural strength and dimensional stability) were achieved with 10% addition.

Generally speaking, a technological feasibility of the biomass ash recycling can be affirmed only in relation to the firing behavior. In fact, the technical constraints of the pre-kiln processes have not been adequately assessed and there may be technological pitfalls, for example about the rheological behavior of slips. Anyway, the literature agrees that the use of biomass ash can lead to properties matching the standards for the BI<sub>a</sub> group or porcelain stoneware tiles, with a recommended recyclable amount in most cases between 5 and 10%. Analogous indications came out for stoneware and wall tiles.

#### ***F. Wastes from metallurgical, steel casting and metal coating industries***

This category includes mainly slags of various origin. Slag is a generic term referring to all non-metallic co-products resulting from the separation of a metal from its ore. Its physical and chemical properties depend on the metal being produced and on the solidification process used, including the type of furnace and in particular the raw materials. These by-products encompass ferrous metallurgy residues, e.g. blast furnace slags (BFS), electric arc furnace slags (EAFS), and basic oxygen furnace slags (BOFS), as well as non-ferrous metallurgy wastes, like copper, lead or zinc slags. The various types of slags are known to be valuable materials with many different applications, for example the air-cooled blast furnace slags are mainly used as cement replacement (Shi and Qian, 2000; Özbay et al., 2016). In addition, wastes produced from metal casting (spent foundry sand) and metal coating (galvanic sludge) have been considered. Overall, the literature for these waste typologies consists of 35 case-studies describing recycling in the ceramic tiles: principally porcelain stoneware (and similar “triaxial” batches) but red stoneware, split and wall tiles as well.

Apparently, metallurgical wastes were tested without any specific care to pre-treatment, except in the few cases where the preliminary grinding of slags (Furlani et al., 2012, 2013; Lim et al., 2016) or a previous calcination of the sludge (Reinosa et al., 2010) were carried out.

On the basis of their physical, chemical, and mineralogical composition, the slags were mainly used either in replacement of clay in simple binary mixes or in substitution of feldspar as a fluxing agent in porcelain stoneware tiles. Some experiments were addressed to novel bodies, mostly made up of waste or mixing raw materials under a rationale different from current industrial tile production.

In the literature examined, the industrial conditions adopted in tile-making were seldom followed in the various manufacturing stages, as

exemplified by wide ranges in terms of pressure in the forming phase (8–100 MPa), firing maximum temperature (up to 1250–1330°C) or schedules lasting many hours. Moreover, all the ceramic products were tested at a laboratory scale and only in a very few cases a comparison with the technical performance of commercial tiles was performed.

The main technological properties of the finished products (water absorption, porosity, linear shrinkage) were always studied; the same does not apply to the performance of semi-finished products for which detailed characterizations have not emerged, besides the importance to outline the influence of waste during all processing steps (e.g., rheological behaviour of the slip, compaction and drying behavior).

*F1. Blast furnace slags* are the most common by-product of pig iron production, discharged from blast furnaces in the primary melting process of iron ore. BFS were experimented under different strategies: in porcelain stoneware (12–30%) as a substitute of feldspar (Dana and Das, 2002, 2003) even mixed with BOFS (Pal and Das, 2013; Siddiqui et al., 2014; Zhao et al., 2015; Pal et al., 2016) or iron ore tailings and/or coal fly ash (Das et al., 1996; Dana et al., 2005). Used also in replacement of limestone and kaolin (6–33%) in wall tiles (Ozturk and Gultekin, 2015) and admixed from 50 to 100% with kaolin (Mostafa et al., 2010).

No data exist about the BFS behavior prior firing. Results of firing tests are contradictory: if a fluxing effect of blast furnace slags (reducing water absorption and increasing shrinkage and MoR) was found in complex batches (Dana et al., 2005) and porous tiles (Ozturk and Gultekin, 2015), the opposite appeared in porcelain stoneware (Dana and Das, 2003; Pal et al., 2016).

Although the recommended recyclable amounts are quite optimistic (30–50% according to Mostafa et al., 2010 and Pal et al., 2016) this picture does not allow any generalized conclusion about the technological feasibility of

BFS recycling beyond small additions to ceramic tile batches.

*F2. Basic oxygen furnace slags* from steelmaking were proposed as fluxing agent in porcelain stoneware and triaxial porcelain batches (6–40%) in replacement of feldspar (Das et al., 2013; Zhao et al., 2014) and even mixed with BFS (Pal and Das, 2013; Siddiqui et al., 2014; Zhao et al., 2015; Pal et al., 2016) or coal fly ash (Das et al., 2013).

Technological data are available only for the firing stage but are often referred to batches based on more than one waste. In general, BOFS tend to anticipate the vitrification (<1200°C) due to the strong fluxing action of the alkali-earths and iron oxide, leading to the formation of a large amount of glassy phase during firing (Das et al., 2013; Pal et al., 2016). The high concentration of CaO in BOFS caused the abundant crystallization of anorthite, at expenses of mullite and quartz. Such phase composition and microstructural changes improved the mechanical strength of the final products. As said for BFS, the available information is too scarce to permit any conclusive statement about the technological feasibility of BOFS recycling in ceramic tiles.

*F3. Electric arc furnace slags* are dense and tough aggregates, which typically contain high amounts of FeO, CaO, and MgO. In addition, small amounts of MnO and Cr<sub>2</sub>O<sub>3</sub> can be present, along with traces of Ba, V, Pb, and Mo, that may constitute a matter of concern for recycling (Quijorna et al., 2014; Mombelli et al., 2016). These features of EAFS require a preliminary treatment of the slag to crush the aggregates and remove metallic iron. Despite their peculiar composition, EAFS have been mainly suggested as fluxing agent in slag-based stoneware batches (10–60%) either substituting feldspar (Dana and Das, 2002; Hasmaliza et al., 2014; Teo et al., 2014; Lim et al., 2016) or clay (Teo et al., 2019a, 2019b). At variance, EAFS were tested (30–40%) in a red stoneware batch (Sarkar et al., 2010) or added to red clays (3–40%) for split tiles (Karayannis et al., 2017).



Grindability was in some cases investigated, denoting a crucial effect of comminution to improve the firing behavior of EAFS-bearing bodies (Lim et al., 2016; Teo et al., 2019a). No further information on the technological behavior prior firing is available. On the other hand, it was demonstrated that slag-based bodies can have excellent technological properties, matching standard requirements for porcelain stoneware tiles in terms of water absorption and mechanical resistance (Sarkar et al., 2010; Teo et al., 2019b). In these bodies, the slag fosters the formation during sintering of both a glassy phase and crystalline compounds, such as anorthite and wollastonite, which are believed to be ensure the chemical and physical properties of finished tiles (Teo et al., 2014, 2019b). Leaching tests corroborate the hypothesis that firing reactions could stabilize EAFS, firmly retaining heavy metals, even though some release may concern chromium (Teo et al., 2014, 2019b).

The above-mentioned studies were carried out at the lab scale, frequently in conditions far from industrial tile manufacturing. This circumstance suggests some caution considering the EAFS amount (40–50%) that is recommended in peculiar slag-based bodies. Although these results are undoubtedly promising, many questions are still unanswered about the EAFS behavior during tile processing. Perhaps, it is not a case that the study carried out in conditions closer to the industrial practice indicates no more than 6% EAFS as the best to recycle in split tiles (Karayannis et al., 2017).

*F4. Non ferrous slags* are produced by metallurgy of copper or zinc. A limited amount of research was conducted on the use of these slags as raw materials for ceramic tile bodies (Abdrakhimov et al., 1990; Naga and El-Maghraby, 2002) or glazes (Ozturk et al., 2019). Anyway, the slags were tested in replacement of clay (10–20%) or dolomite (up to 30%) with conventional processing and sintering steps. Lab scale trials show an accentuated fluxing effect leading to good technical performances within the category of red stoneware or split tiles, like

water absorption  $\geq 3\%$  and MoR around 40 MPa (Naga and El-Maghraby, 2002). The precautions already expressed regarding the technological feasibility of metallurgical slags are even more valid in this case, due to the limited number of available data.

*F5. Spent foundry sands* are utilized together with binders (e.g., bentonite) to create molds or cores, particularly for casting iron-based alloys. Although foundries generally use high quality quartz or zircon sands, olivine sand is employed for the production of manganese containing steel parts to prevent undesired effects between quartz particles and the metal under casting. Spent sands are not always recovered separating each type, so the composition may significantly vary according to the mix. Moreover, spent sands usually retain debris of cast metals and their impurities, such as Pb, As, Cr, Mo, Ni, Cu, Hg (Alonso-Santurde et al., 2010; Siddique et al., 2010). Quartz sand was essentially experimented in clay:waste binary mixes, up to 50% in red clay (Quaranta et al., 2010; Coronado et al., 2015; Lin et al., 2018) or up to 60% in ball clay (Luo et al., 2014). In addition, it was an ingredient of multiwaste bodies, including glass cullet, metallurgical slag and/or sewage sludge ash (Coronado et al., 2015; Lin et al., 2017). Similarly, olivine sands were admixed (20–60%) with different red clays (Furlani et al., 2012, 2013).

Since foundry sands present a grain size coarser than common ceramic raw materials, they were used after milling to avoid an increase of porosity in the final products. The above-mentioned studies concerned only the firing stage, by which it was demonstrated that clay bodies containing 10–15% quartz sand (after firing at 1100°C) or 40% olivine sand (fired <1200°C) gave good technological performances for floor tiles (Quaranta et al., 2010; Luo et al., 2014; Lin et al., 2018) or split tiles (Furlani et al., 2012). However, there are clues of black core problems (Lin et al., 2018) and release of toxic elements, like Cr or Mo (Coronado et al., 2015), which along with the

lack of information on many tile-making stages push for caution about the technological feasibility of this waste recycling.

*F6. Galvanic sludges* stem from cleaning of wastewater resulting after chemical treatment of metallic surfaces. Their chemical composition is reknown for a high concentration of hazardous elements, such as Cr, Zn, Ni, Cu, and Pb (Karlovic et al., 2008; Ozdemir and Piskin, 2012). Moreover, they show the presence of crystalline phases, such as quartz, calcite, and gypsum. This implies a certain amount of gas-forming species, so the sludge was used only after a pre-calcination at 1125°C for 1 h. After this heat treatment, the ash contained spinel ( $\text{NiCr}_2\text{O}_4$ ) and diopside ( $\text{CaMgSi}_2\text{O}_6$ ). Only three studies considered this kind of waste as ingredient in stoneware tiles up to 20% (Ferreira et al., 1999; Silva et al., 2010) or 20% together with glass cullet (Reinosa et al., 2010). The resulting products exhibit a porosity and mechanical properties similar to those of commercial tiles, so fulfilling the standard requirements. Most important, the release of Cu, Cr, Ni and Zn was extremely low, even though leaching was tested in hot water (Reinosa et al., 2010). However, a disadvantage is the dark colour of the finished product. Although encouraging, these achievements where at the lab scale only and the lack of information about crucial steps in the ash or sludge recycling (e.g., rheological behavior of slip) suggest extreme caution about the feasibility of galvanic sludge reuse in ceramic tiles.

### **G. Wastes from the production of ceramics, glasses, and refractories**

The ceramic tile factories have since long time developed a range of technological solutions to cannibalistically recycle almost all wastes deriving from the different phases of the production process (Timellini et al., 1983). Nowadays, most of the residues produced by the ceramic tile industry can be reused, as demonstrated by the pluriannual experience of waste management in the Sassuolo district in

Italy (Busani et al., 1995; Palmonari and Timellini, 2000). However, there is a gap between the actual industrial use of ceramic residues and the technological knowledge about the effect of the different components on the process and product parameters. In the industrial practice, waste recycling is managed empirically, relying more on trial-and-error than on a design of experiment, and results are just occasionally divulged. In any case, this cannibalistic loop is made possible by two main reasons: the quite constant composition of wastes and a composition fully compatible with that of the desired product. The main ceramic residues stem from: purification of waste waters, including glazing sludges; unfired (green) scraps and dust; scraps of fired products, including waste frit; tile polishing; spent lime of kiln scrubbers.

*G1. Ceramic wastewaters and sludges:* apart losses due to evaporation during drying processes, water used in wet milling is recycled (about the 70% of the need) through a closed loop, involving the whole process (washing and cleaning operations, glaze preparation, condensation in the various driers). All these wastewaters are collected and decanted, in order to separate the suspended solids as a sludge, prior undergoing several cleaning treatments (Manfredini et al., 1991; Zannini, 2020). The continuous recycling of waste waters, however, leads to a progressive increase of dissolved ions, coming also from cleaning additives, which can modify the slip rheology, and consequently the milling behavior. Also, aesthetic properties of final products and gaseous emissions during firing may be affected (Zannini, 2020). At all events, the direct use of waste waters, without settling, in the wet grinding process may lead to economic benefits (Manfredini et al., 1991). Nevertheless, a progressive substitution of decanted water by the wastewater as collected led to a slip viscosity boosting attributable to the increase of both solid content and soluble Mg and Ca (Andreola et al., 2004). The sludge obtained from water purification can be added

directly in the mill. In general, the slip viscosity is linearly correlated with the sludge addition and a content up to 5% is compatible with the industrial requirements (Manfredini et al., 1991). In alternative, the sludge, once previously dried, was tested in substitution (up to 50%) of the batch for porous tiles (Amin et al., 2019). The sludge addition slightly increased the body plasticity, as a result of the higher content of clay minerals. At the same time, a strong decrease of bending strength was observed. This effect is more evident in the case of contemporary presence of other green and fired ceramics wastes, even if the effect on the firing behavior is not well defined (Elmahgary et al., 2018). The addition of glaze sludge promotes the sintering process by reducing the firing temperature, besides a content higher than 5 wt% should be avoided (García-Ten et al., 2015; Amin et al., 2019). When a sludge from the glazing process was added to a stoneware batch up to the complete replacement of feldspar, firing shrinkage, bulk density and bending strength were increased, while plagioclase crystallized during firing, favored by the higher amount of alkalis and alkali-earths (Wannagon et al., 2012; Theerapapvisetpong and Nilpairach, 2015).

*G2. Ceramic green scraps* include wastes produced before the firing process (broken tiles, slip and powder sieving rejects, dust). These residues exhibit different physical status and particle size distribution: broken tiles are made up of irregular scraps or lumps; slip sieving after milling provides a mud; powder sieving after spray-drying gives clumpy rejects. Broken tiles must be previously crushed but can be reused without any further milling (as powder rejects and dust) even if added directly to the slip. Rejects of slip sieving are put again into the mill. In any case, mandatory is that the waste introduction, either in the wet milling process or before spray-drying, must not induce any intolerable changes to slip rheology. Although this is a widespread industrial practice, more than for other kinds of waste, no scientific

papers can be found, apart a discussion in multi-waste studies (García-Ten et al., 2015; Rambaldi et al., 2016, 2018; Valença and Ferrazo, 2018). In general, the introduction of smaller particles into the mill reduces the required milling time/energy. At the same time, a decrease of firing temperature can be assumed, given to a finer particle size of the waste components milled twice. Green scraps act as plastic materials, increasing the compactness and the mechanical strength of the unfired tiles (García-Ten et al., 2015). From the scarce results reported in the literature, a waste recycling up to 50 wt% seem to be technologically feasible for all tiles formulations. However, the physiological loop of green scraps can be inferred from data about the Italian tile district: 10–15% of the whole production (Resca et al., 2015).

Dust from spray-drying air purification is produced from the cyclone that follows the spray drier. Given by the specific origin, it is characterized by a smaller particle size with respect to standard spray-dried powders (El-Fadaly et al., 2010; Amin et al., 2019). Similarly to green scraps, the dust recycling is more popular in the industrial production than what foreseeable by academic results. Papers dealing with this waste are in fact scarce.

The effect of dust addition on plasticity is not well defined and depends on the specific formulation tested. Plasticity, as assessed by the Pfefferkorn method, increased of ~22% for a substitution of 50% in a batch for wall tiles, but the same substitution in a partially vitrified floor tile mixture did not modify substantially the plasticity. In both cases, the dust recycling brought about a higher green bulk density (Amin et al., 2019). Effect on the drying stage cannot be generalized, since no clear trend was observed for shrinkage (Amin et al., 2019) or, on the contrary, an increase of about 50% was recorded for 10% of dust addition (Elmahgary et al., 2018). In porous formulations, a small dust content dramatically reduced the green modulus of rupture (MoR), while in floor tiles a 5–10%

of dust doubled the mechanical strength (Elmahgary et al., 2018; Amin et al., 2019). Dust-containing batches were tested simulating industrial fast firing conditions, in the temperature range between 1000 and 1150°C. The properties of fired tiles were not significantly affected, showing similar values up to a waste content of 40% (El-Fadaly et al., 2010; Amin et al., 2019). Also, mechanical strength matched the standard prescription, testifying a good compatibility of the waste with light-firing stoneware. However, the contemporary addition of dust (5%) and wastewater sludge (30%) worsened the product performances (Elmahgary et al., 2018). Otherwise, in case of porous wall tiles, waste addition led to a moderate deterioration of properties: increasing water absorption and decreasing bending strength (Amin et al., 2019). Therefore, it can be concluded that dust recycling is technologically feasible up to a maximum waste content of ~20%.

*G3. Scraps of fired ceramics* represent a composite group in terms of both ceramic material (wall and floor tiles, sanitaryware, refractories, waste frit) and process generating the waste. They are originated during tile finishing (cutting, grinding, and squaring) and product sorting (tiles broken or discarded for intolerable flaws) or by stock management (unsold lots). As a consequence, their compositions faithfully reflect that of the recycled ceramic material, apart residues of abrasive tools and glaze coatings. Fired scraps must be crushed before being reused, in order to match the usual grain size of raw materials they can substitute: filler and/or flux (Karamanov et al., 2006; Oluseyi and Das, 2014; Tarhan et al., 2017; Kabiraj et al., 2018). In fact, this kind of wastes is commonly used as skeleton, given to their lower reactivity, since already fired, instead of hard components. The incorporation of fired scraps into porcelain stoneware tiles (or other porcelain-like products) is quite described in the literature. The complete substitution of quartz with porcelain

scraps (20%) in a standard porcelain formulation led to the same particle size distribution of the reference body, with no apparent effect on the milling performance (Kabiraj et al., 2018). Overall, the resulting particle size is strongly dependent on the milling technique (García-Ten et al., 2015) as the consequent effect on dry mechanical strength, which can be either positive (Kabiraj et al., 2018) or more frequently negative (García-Ten et al., 2015; Valença and Ferranço, 2018; Elmahgary et al., 2018).

In a porcelain batch, the addition of fired scraps up to 20% led to an increased firing shrinkage in both slow and industrial-like schedules (Karamanov et al., 2006; Tarhan et al., 2017; Kabiraj et al., 2018). This difference is conspicuous in case of quartz replacement, indicating a stronger contribution to densification given by the higher alkali amount of scraps (Kabiraj et al., 2018). On the contrary, for a partial substitution of both quartz and feldspar at slow heating rates, the firing behavior is significantly affected by the waste only beyond 1300°C (Karamanov et al., 2006). In both cases, the maximum sintering temperature was lowered in presence of fired scraps. In any case, water absorption was reduced, and mechanical strength was improved because of waste addition. Moreover, when sanitaryware scraps substituted quartz, the thermal expansion coefficient went down, improving dimensional stability (Tarhan et al., 2017). Finally, refractory roller scraps were used (2%) in porous tile production with a limited effect on technological properties (Elmahgary et al., 2018). In general, the complete substitution of filler, and in part of fluxes, in porcelain stoneware batches with ceramic scraps is perfectly feasible, allowing to maintain or even to improve the technological properties. On the contrary, the addition of waste fireclay (sanitaryware) up to 15% in place of kaolin for monoporosa production is not recommended, because of a limited worsening of technological properties (Tarhan et al., 2016).

Residues generated during frit manufacturing are seldom recycled in ceramic tiles (García-Ten et al., 2015). Their chemical composition is usually rich in alkalis, alkali-earths, and even strong fluxes, like B and Zn. For this reason, their addition to vitrified tiles favors sintering and permits a reduction of firing temperature. However, no data are available about the feasibility of recycling, due to the lack of papers discussing the effect of this kind of wastes on the technological properties.

*G4. Sludge from ceramic tile polishing* is a byproduct of surface polishing operations aimed at improving the tile aesthetics (providing brightness and mirror glass effect) and turning the final product similar to natural stones. As a result, wastewater with both ceramic dust and fragments of abrasive tools constitute the polishing sludge, which is obtained by dewatering to a semisolid consistence. Usual amounts of 1-5% of abrasive (SiC) and 2-6% of binder (magnesium oxychloride) are declared (Shui et al., 2011) but there is a remarkable variability upon the surface treatment: from deep polishing to mild lapping. The latter is currently carried out with diamond tools with a minimal mass removal. Polishing sludge was tested as addition (up to 80%) to the mill, either in place of feldspars or of the whole batch for porcelain stoneware tiles. As SiC particles have generally a size between 5 and 10  $\mu\text{m}$ , no granulometric mismatch with the porcelain stoneware slip is expected (Ke et al., 2016). During the milling step, a sludge addition up to 20% did not modify the slip particle size distribution but led to an increased bending strength of green tiles (Kabiraj et al., 2018). However, the polishing residues bring about a major drawback related to the occurrence of silicon carbide. In fact, the SiC oxidation, occurring during firing, leads to the formation of CO and CO<sub>2</sub>, and consequently to a considerable bloating of tiles (Xi et al., 2012; Ke et al., 2016) with permanent deformations and black core. The sludge exhibits by itself a propensity to expand already at 1040-1060°C

(Xi et al., 2012; Ke et al., 2016). The individual presence of silicon carbide or magnesium oxychloride, up to 2% into the porcelain stoneware mix, provoked a decrease of bulk density of about 20%; such an effect was dramatically boosted by their concomitance, leading to a density drop of about 50% (Xi et al., 2012). Nevertheless, once used in place of sodic feldspar (5-15%) in porcelain stoneware, it lowered water absorption and firing temperature down to 1140°C, achieving a bulk density of  $\sim 2.41 \text{ g/cm}^3$  and a flexural strength of  $\sim 97 \text{ MPa}$ . The incipient body expansion – observed at 1160°C or 1140°C for 15% sludge – constrains both firing schedule and amount of waste recyclable (Rambaldi et al., 2007a). A similar effect on mechanical properties was observed when the sludge was used (20%) instead of quartz in porcelain (Kabiraj et al., 2018). In all cases, the larger alkali amount promoted the mullite formation and a structural reinforcement is envisaged for well-crystallized mullite needles grown in the feldspathic glass. Anyhow, the strong tendency to bloating makes problematic any direct utilization of the polishing sludge in vitrified tiles. No industrial recycling, at best of our knowledge, has been declared.

For this reason, the recycling of this waste was addressed to lightweight products of novel design. In general, a linear correlation between waste content (up to 80%) and bulk density decrease was observed, with an extent that depends on the specific material. For sintered products, like porcelain and stoneware fired between 1160 and 1200°C, a 50 wt% of sludge reduced the bulk density by 70% to 50%, as a function of the different experimental formulations (Xian et al., 2015; Silveira et al., 2014; Ke et al., 2016). For instance, 10% of sludge led to the formation of a larger amount of liquid phase, which can decrease porosity at lower temperature, transforming interconnected pores into spherical ones, larger and more abundant (Xian et al., 2015; Ke et al., 2016). By increasing the waste content, an even more

porous microstructure was achieved, fractured by gas evolution, with a consequent decrease of mechanical properties. A compromise was found for 10% of sludge, which ensured an adequate flexural strength after firing at 1140°C. In these conditions, needle-shaped mullite crystallization occurred already in fast firing at 1120°C with a content increasing with the sludge amount (Xian et al., 2015; Ke et al., 2016).

*G5. Spent lime* produced by flue gas scrubbers of roller kilns is a hazardous waste. During the firing process, several volatile molecules are evolved, coming from both raw materials and additives. Chlorine, fluorine and sulfur acids and compounds, which are released within the kiln, are subjected to stringent regulations, which make mandatory the presence of filtration systems able to retain them. These filters are based on powders or lumps of basic substances, generally lime, which selectively react with some contaminants present in flue gases, thus blocking them. These filters generate a solid waste (spent lime) which is currently landfilled. The recycling of this spent lime within the same ceramic tile factory would represent an obvious pathway. As the waste occurs in the form of powder, it can be easily introduced in the milling step (Andreola et al., 1992a, 1992b). This recycling solution was practiced during 1970s and 1980s, adding the waste to ceramic batches fired at relatively low temperatures, first empirically, then demonstrating, with tests performed under industrial conditions, that the spent lime recycling was technologically feasible up to 1.2% in almost all tile formulations (Andreola et al., 1993). Nonetheless, it was gradually abandoned with the conversion to porcelain stoneware, when it emerged that the spent lime could have an environmental impact given by fluorine mobilization during firing for temperatures higher than ~1000°C, depending on firing cycle (Dondi et al., 1990). In addition, chlorine can be even more easily released during the firing process, increasing its content into vent streams.

For these reasons, the recent efforts to include the spent lime in optimized batches, designed to get 100% waste porcelain stoneware tiles (Garcia-Ten et al., 2015; Rambaldi et al., 2016; Rambaldi et al., 2018) had no industrial repercussion. In detail, there are several technological hindrances to spent lime recycling. For example, given the high content of soluble calcium, a waste addition during milling strongly increases the slip viscosity, requiring higher amounts of deflocculant and/or water to correctly manage grinding and spray-drying operations. The rheological behavior was tested, up to 1.6% waste, in both porcelain stoneware and monoporosa slips (Andreola et al., 1992a). The effect is strongly dependent on the slip and the deflocculant nature. In case of porcelain stoneware, the original slip viscosity was maintained, by optimizing deflocculant type and amount, only for spent lime additions lower than 0.3% (Andreola et al., 1992a). Higher concentrations (up 0.5%) did not modify the properties of fired tiles, but with a waste percent as low as 4 wt% important deformations were observed (Andreola et al., 1992b). For intermediate concentrations, porcelain stoneware showed a lower firing shrinkage and a higher water absorption, as a function of waste addition, suggesting a clear interference in the standard sintering process. At variance, an opposite effect was observed in porous glazed wall tiles, where spent lime favored the densification. These trends were observed for both slow and fast industrial-like firing schedules and were associated to an increasing anorthite crystallization at expenses of mullite, turning important for waste higher than 4% (Andreola et al., 1992b).

#### ***H. Wastes from construction, demolition, and dredging***

These waste sources attracted a limited attention as potential supply of the ceramic tile production. Most of the research efforts were spent to solve two environmental problems, entailing the disposal of sediments from dredging harbors, rivers, and estuaries (often

contaminated) or construction and demolition wastes (CDW) and particularly asbestos-containing residues. Interestingly, research was carried out not only at the laboratory level, but the scale-up to a commercial secondary raw material was achieved in the case of CDW.

*H1. Construction and demolition wastes* include common residues – i.e., mixture of brick, stone, concrete, and mortars in variable ratios – that were experimented by mixing with a red clay (Acchar et al., 2013b). CDW addition up to 10% had no significant changes in the final properties, paving the way for a possible use in porous tiles. In contrast, waste at ~20% modified its technological role from inert to flux, so shifting the behavior of the body towards vitrified floor tiles. In any case, the outstanding variability of CDW in both composition and particle size distribution (Bianchini et al., 2005; Rodrigues et al., 2013) represent a key point that does not allow any general conclusion about technological feasibility of recycling in ceramic tiles. Another question is the possible mobilization of heavy metals, such as Cr, Se, Mo, Cu, S, Cl (Delay et al., 2007; Butera et al., 2014). In order to overcome this hindrance, the importance of selective waste collection is stressed (Acchar et al., 2013b). This is the starting point in the treatment of hazardous demolition residues, essentially cement-asbestos slates (Leonelli et al., 2006; Gualtieri et al., 2008). Given the known carcinogenic hazard, asbestos must be completely transformed into inert silicate phases, e.g. by melting to a silicate glass at  $T > 1250^{\circ}\text{C}$  (Gualtieri et al., 2008) or by microwave inertization (Leonelli et al., 2006). By such preliminary treatment, asbestos was transformed into harmless forsterite. Laboratory trials with porcelain stoneware batches containing the transformed asbestos residue (5%) exhibit a lower water absorption and a higher bulk density (Gualtieri et al., 2008) or no significant variation with respect to the waste-free body (Leonelli et al., 2006). Such promising results fostered the scale up to the industrial production of a secondary raw material (KRY

AS) obtained by the thermal transformation of cement-asbestos into properly designed glass-ceramic frits. KRY AS addition of 5% to porcelain stoneware slabs resulted into comparable or even improved technological properties: better stain resistance, lower firing shrinkage, improved mechanical strength achieved for a water absorption  $< 0.5\%$ , using the firing schedule optimized for the waste-free body (Ligabue et al., 2020).

Cement kiln dust is a fine-grained residue rich in CaO, silica and alumina. It was tested (up to 15%) in a wall tile batch in substitution of calcite (Aydin et al., 2019). As a result, open porosity and water absorption increased, while bulk density decreased, with the amount of waste. The crystallization of anorthite and diopside, induced by the dust addition, promoted a beneficial decrease of moisture expansion and a whiter color of finished body. Recommended amount of cement kiln dust is up to 3 % in porous tile bodies.

*H2. Dredging soils* resulting from river or harbor dredging are sediments with a composition approaching that of clay raw materials. Nonetheless, they are considered hazardous materials because of possible contamination by heavy metals, such as As, Cd, Cr, Cu, Pb, V and Zn (Tack et al., 1999; Borma et al., 2003) as well as organic substances (e.g., hydrocarbons). Relevant from the technological point of view are the relatively high amount of iron oxide and moisture (semi-solid wastes). On the other hand, the modest concentrations of S and Cl may be matter of concern for volatile emissions. Sludges stemming from purification of drinking water are instead particularly rich in aluminium and iron oxides. Dredging spoils were investigated as ingredients to produce ceramic tiles: alone (Baruzzo et al., 2006; Romero et al., 2008) or up to 20% in replacement of quartz (Hossain et al., 2019) or clay materials (Torres et al., 2009). Analogous trials were run using the sediment (up to 65%) from water supply treatment (Junkes et al., 2012; Wangraddiskul et al., 2017; Rodrigues and

Holanda 2018). Batches made up of waste only exhibit many defects (too high firing shrinkage, tendency to overfiring with increase of closed porosity) but also promising results (good mechanical properties, retention of many hazardous metals). Hence, the investigated wastes were considered as suitable secondary raw materials in ceramic tiles (Baruzzo et al., 2006; Romero et al., 2008). The use of river silt in replacement of quartz resulted, at a firing temperature of 1130°C, in a slight increase of water absorption and decrease of shrinkage, with no significant change in the mechanical properties of floor tiles (Hossain et al., 2019). At variance, when the river silt was introduced instead of clay, final properties (bending strength, bulk density, firing shrinkage and water absorption) were similar to those of the standard industrial body. The recommended addition is up to 5% (Torres et al., 2009). The technological behavior of the waste was considered suitable also in substitution of kaolin, leading to higher bending strength, lower water absorption and higher firing shrinkage. In this case, the recommended recyclable amount of sediment is 15% (Wangrakdiskul et al., 2017).

### ***I. Wastes from municipal wastewater treatment plants***

Municipal water treatment and purification process are importantly rising all around the world, given by both populations growing and even more severe environmental restrictions on waste disposal. Furthermore, water treatment processes are strategical in geographical areas with limited water resources. A large number of sludges are obtained from water treatment plants as byproducts (Municipal Sewage Sludge - MSS), as many as the different process stages (Świerczek et al., 2018). Nowadays, three main disposal pathways are used: land or sea dumping, recycling as agriculture fertilizer or as incinerator fuel (Świerczek et al., 2018). The direct input in soils has a negative impact on people and environment health. At the same time, MSS handling is a real problem and the possibility of their utilization in ceramics production has been

extensively studied in latest years (Chang et al., 2020). The main issue related to the use of MSS is the huge water content. It is about a semi-liquid waste, mainly composed by water up to 95–98% (Chang et al., 2020). For this reason, MSS is firstly pre-dried and then reused and recycled. This pretreatment requires a high energy consumption, limiting the application of MSS at industrial scale. MSS composition is generally quite variable, depending on the geographical origin, the proximity to industrial sites, the period of the year and the technology used for the wastewater treatment. All these parameters affect amount and nature of the inorganic (Sommers, 1977) and organic sludge compounds (Dondi et al., 1997). Furthermore, the specific steps involved in the purifying process increase ferric or aluminum salts (used to favor the flocculation process) and chlorine derivatives. More importantly, the effluent origin influences the presence of toxic elements (Xu et al., 2013; Yang et al., 2013) and potential biohazards (i.e. microorganism, pathogens (Giger et al., 1984; Giller et al., 1998; Gerba and Smith, 2005; Ruan et al., 2012). Sewage sludges tested in ceramic tiles exhibit significant concentrations of heavy metals, namely Cr, Co, Cu, Ni, Sb, Sn and Zn (Carlson and Morrison, 1992; Fjällborg et al., 2005) along with rather high amounts of P, S and Cl.

Several studies about the possibility to use MSS for building materials production have been carried out: cement (Fytili and Zabaniotou 2008; Rodríguez et al., 2010), bricks (Dondi et al., 1997; Weng et al., 2003; Liew et al., 2004), ceramic pellets (Watanabe and Tanaka, 1999; Camilla and Jordán, 2009). However, only few examples can be found for tiles production (Chang et al., 2020). By the way, considering the large amount of raw materials used and the market share of the final products, the ceramic tile industry seems to be the most attractive sector to absorb MSS wastes. Very few papers report data related to the effect of MSS on the technological behavior of bodies, even less for sintered products, but a quite wide range of



concentration (1–65%) was studied (Abadir et al., 2004; Jordán et al., 2005; Montero et al., 2009b; Jordán, 2010; Amin et al., 2018).

An important gap about the effect of MSS addition on grindability or slip rheology is still to be covered by literature results. At the same time, only few data are available about its effect on the other technological properties. Generally, a quite linear relation between the MSS content and the obtained plasticity can be found. Amin et al. (2018) reported that plasticity doubled in the waste range between 0 and 35%, observing an increase of the plasticity number from  $\sim 25$  to  $\sim 50$  (Pfefferkorn method). Similarly, but in a smaller range of waste content (0–10%), an increase of the plasticity index (11.3–12.5) and both plasticity limits (UPL 31.8–35.8%; LPL 20.5–22.8%) was observed (Rodrigues and Holanda 2018). This effect can be correlated to both the lower particle size of the sludge (Amin et al., 2018) and the presence of organic matter (Abadir et al., 2004). On the contrary, the increased MSS content led to a progressive decrease of the MoR for the dried tiles (Jordán et al., 2005): for instance, the increase of MSS till 10% generated a MoR reduction of about 30% (Amin et al., 2018). The contemporary addition of marble residues emphasizes the loss in mechanical properties of the green product (Montero et al., 2009b; Jordán, 2010).

The effect of clay substitution by MSS depends importantly on the firing conditions. The rise of sludge content improved firing shrinkage in a wide range of firing temperature (1100–1250°C), with measured values between 5 and 11% (Amin et al., 2018). This phenomenon is more important at firing temperature typical for vitrified products, while becomes less significant at lower temperature, e.g. 1050°C (Jordán et al., 2005; Montero et al., 2009b; Jordán, 2010). This difference can be explained by the presence of iron oxide that reduces the liquid phase viscosity, enhancing sintering (Segadães, 2006). Water absorption underwent different trends, as a function of the temperature range studied. At

lower temperature, between 1050 and 1150°C, the increase of waste content brought about a general increase of water absorption, followed by a strong decrease in the mechanical properties of fired tiles, for the whole waste concentration range. For a standard heating cycle for porous ceramic bodies, the addition of 10% of dried sludge boosted the water absorption of about 37%, which corresponded a drop in MoR of 73% (Jordán et al., 2005). The co-addition of marble residues increased the porosity, maintaining quite constant the MoR (Montero et al., 2009b; Jordán, 2010). Similarly, in the 1050–1150°C range, the addition of 35% of waste almost quadruplicated water absorption (e.g. from 12% to 40% at 1050°C), zeroing the bending strength (Amin et al., 2018). However, increasing firing temperature in an industrial-like rapid cycle (1150–1250°C) with a large addition of crude MSS in a split tile production induced just a limited porosity increment and a quite stable bending strength (Zhou et al., 2013).

Given to the scarcity of information, it is not easy to recommend a waste concentration range to optimize both process and product. A content of MSS up to 10% in the formulation of dense bodies seems to cause no important change in terms of physical properties. Sintering process can considerably reduce the leaching of heavy metals (e.g. Cu, Zn, Cd, Pb), allowing to maintain detected concentration lower than regulatory level required (Favoni et al., 2005; Zhou et al., 2013).

Not least, the smells produced from organic matter decomposition during waste storage and processing, and especially firing, must be kept under control. Environmental problems given by gaseous emissions and odors can be an important hurdle to overcome a massive approval by industries and people. It must be taken into account that waste ceramization leads to lower emissions respect to other treatments, e.g. incineration (Fytli and Zabaniotou, 2008), but at the same time the presence of MSS may strongly rise the content of both VOCs and

inorganic pollutants in the kiln flue gases (Cremades et al., 2018). In particular, the increase of some odorous molecules was observed (e.g. dimethyl disulfide, methyl mercaptan, acetic acid). Strong smells can be detected also given by unburnt organic matter. Associated to the chlorination of water during the treatment process, higher HCl content are measured. The upgrade of the control protocols and air treatment processes, required to minimize direct and indirect side effects, certainly makes more complex and expensive the production.

### ***J. Solid wastes from municipal and industrial sorting***

Solid wastes from municipal and industrial sorting include different types of glass. They derive from the separated collected fractions of: container and flat glass, referred as Soda-Lime-Silica (SLS); glasses from Waste Electrical and Electronic Equipment (WEEE): PC-TV screen, PC-TV cathodic tube (CRT), different types of lamps; other post-consumer glasses (such as thin-film transistor liquid-crystal display TFT-LCD, veal, solar panels); glasses from waste melting (municipal solid waste incineration, dredging spoils, vitrification of various wastes). All of them are amorphous materials, or present a negligible crystalline component, and can be basically distinguished on their chemical composition. The main issue, which prevents the direct use of these glasses in the ceramic process, is related to their physical state, specifically granulometry. The glass particle size, in fact, depends on the waste collection system and following treatments. Usually, it does not match the needs of the tile-making process. This technological gap imposes a proper milling of the waste glasses before their use as ceramic raw materials. Other pre-treatments are related to the presence of pollutants and are type-specific: for the SLS, labels and glue on container require a washing in order to avoid rheological problems to the slips. Likewise, WEEE glasses can present residues of plastics or metal circuits, while phosphorescent lamps or solar panels can

have functional coatings, to be removed. Some concerns on environmental and health problems are related to the presence of hazardous elements, such as Pb, Sr and Ba in cathode-ray tube and panel waste glass (Musson et al., 2000; Méar et al., 2006; Yot and Méar, 2011); As and Sb in LCD (Savvilotidou et al., 2014); Hg in fluorescent lamp waste (Rey-Raap and Gallardo, 2012; Tunsu et al., 2014). They have to be taken into account, particularly in case of obsolete materials, which no longer respond to the current law requirements.

The use of waste glasses in the ceramic tile production has been extensively investigated in the last decades, for their fluxing power able to lower the firing temperature (Silva et al., 2017), as well as for their potential in lower the consumption of natural resources and the costs of waste disposal (Raimondo et al., 2007). Especially, the employ of SLS in substitution of the fluxing component (feldspars) in triaxial porcelain stoneware mixtures, was the object of many papers. The introduction of SLS has been also evaluated in mixtures for dark-firing bodies (vitrified, porous and split tiles), and waste-based bodies too, providing a variegated picture regarding the employment of this type of glass. However, despite the large amount of literature (almost 40 articles revised), less than half report significant technological data. The majority of these works followed lab scale processing conditions far from that of the ceramic industry, with quite slow firing cycles.

Anyway, some example of industrial-like cycle at lab scale (Matteucci et al., 2002; Pontikes et al., 2005; Faria and Holanda, 2016), tests on a pilot line (Carbonchi et al., 2003; Tucci et al., 2004) and even a prototype production (Skerratt, 2001), were found. For as concern all the other glasses, their use was much less examined. The available literature consists of 24 papers in total, some dealing with both PC-TV screen and CRT (Andreola et al., 2005; Tucci et al., 2006; Raimondo et al., 2007; Bernardo et al., 2009). Just twelve present significant technological data, relative to: glass wastes in

substitution of feldspars in porcelain stoneware bodies or clay in dark-firing bodies (both vitrified and porous). Along with regular pressing conditions and fast-firing (Brusatin et al., 2005; Raimondo et al., 2007, Andreola et al., 2008, 2010; Chemani, 2012), sometimes the firing conditions adopted do not match current technologies in tile-making (Rambaldi et al. 2004; Tucci et al., 2006; Lin, 2007; Bernardo et al., 2008a, 2008b; Morais et al., 2011; Kim et al., 2016).

*J1. Soda-Lime-Silica glass.* At variance of fluxes utilized in the ceramic tile industry, such as feldspars, SLS shows lower contents of alumina and higher levels of soda and lime, resulting more fusible. For this reason, SLS has been mainly employed as flux in substitution of feldspars (up to 30%) in porcelain stoneware batches (Matteucci et al., 2002; Carbonchi et al., 2003; Rambaldi et al., 2004, 2007; Tucci et al., 2004, 2006; Bakr, 2005; Pontikes et al., 2005; Tarvornpanich et al., 2005; Rambaldi et al., 2007b; Luz and Ribeiro, 2007; Viruthagiri et al., 2009; Fraga et al., 2011; Mustafi et al., 2012; Ajanaku et al., 2015; Chitwaree et al., 2018). Nevertheless, other strategies of incorporation were tested, e.g., in substitution of clay (up to 50%) mostly for red stoneware (Theerapapvisetpong and Nilpairach, 2015; Njindam et al., 2018), with one example up to 5% for porous tiles (Faria and Holanda, 2016). Otherwise, trials were run with glass in replacement of the whole body (up to 50%) in red stoneware (Youssef et al., 1998), porous (Youssef et al., 1998; Darweesh, 2019) or split tiles (Pontikes et al., 2005, 2007b). Waste-based products were also investigated, such as SLS mixed with cockle shells in proportion from 50:50 to 70:30 (Shamsudin et al., 2018) or 95% SLS plus additives (Skerratt, 2001). Unfortunately, in both papers, data are insufficient to evaluate the technological performances of finished products.

The effects of SLS glass on the technological behavior prior firing was rarely investigated, not

showing particular deviation from the benchmark in terms of drying shrinkage for porous tiles (Faria and Holanda, 2016), dry bending strength for porcelain stoneware (Matteucci et al., 2002) and plasticity for split tiles (Pontikes et al., 2005). Otherwise, the technological properties of the fired batches describe specific trends, reflecting the different incorporation strategies and therefore the tile type. SLS in substitution of flux in porcelain stoneware does not significantly affect the firing shrinkage and water absorption, even up to 20% of addition; on the contrary, a worsening of the bending strength can be observed also at low SLS concentration (Fig. E). Moreover, further experimental data indicate for batches containing SLS >10wt%, an increasing of closed porosity, a loss of bulk density (Matteucci et al., 2002; Tucci et al., 2004; Mustafi et al., 2012) and even crystallization of undesired phases (such as plagioclase in case of high Ca concentration), which inhibited the mullite formation (Pontikes et al., 2005). For these reasons, a recommended SLS waste amount in porcelain stoneware is in the 5–10% range. In contrast, the introduction of SLS glass in replacement of clay (for red stoneware) or the whole body (for porous and split tiles) induced a different behavior. It increased the firing shrinkage, lowering the water absorption and improving the bending strength (Fig. E). It can be said that in these batches the glass can act both as a flux, promoting the sintering and the closure of open porosity – resulting in a lower water absorption – but also as a filler, at least at relatively low temperatures. Suitable properties were found for all these tile types, with a recommended waste amount in the 10–30% range.

*J2. PC-TV screen glass* represents the front part of television and computer monitors with a chemical composition enriched in alkali, barium and usually strontium. Barium released by leaching from the as-received waste glass is in very low amounts (Andreola et al., 2008).

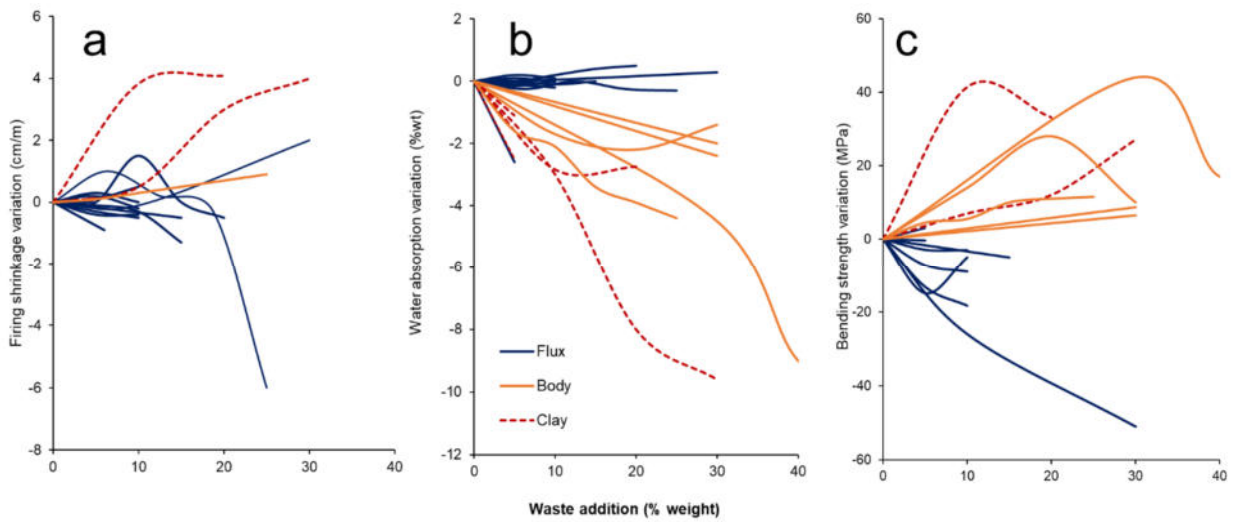


Figure E. Effect of waste glasses on the firing behavior of ceramic tile bodies. Variation of: a) linear shrinkage, b) water absorption, c) bending strength, as a function of waste amount. Flux (Matteucci et al., 2002; Carbonchi et al., 2003; Rambaldi et al., 2004; Tucci et al., 2004; Bakr, 2005; Pontikes et al., 2005; Tarvornpanich et al., 2005; Tucci et al., 2006; Rambaldi et al., 2007b; Mustafi et al., 2012; Ajanaku et al., 2015). Body (Youssef et al., 1998; Pontikes et al., 2005; Darweesh, 2019). Clay (Theerapavisetpong and Nilpairach, 2015; Faria et al., 2016; Njindam et al., 2018).

PC-TV glass has been investigated in substitution of feldspars (up to 35%) in porcelain stoneware (Tucci et al., 2006; Raimondo et al., 2007, Andreola et al., 2008) and in place of clays (up to 20%) in red stoneware (Andreola et al., 2010). As in the case of SLS, information about the behavior of the unfired mixtures is scarce. A slight increasing of the dry MoR and values comparable to the benchmark for drying shrinkage and dry bulk density were found in porcelain stoneware with glass up to 10% (Raimondo et al., 2007). The effect of this glass during firing substantially replicates that of SLS in porcelain stoneware: firing shrinkage and water absorption stood rather constant with up to 10% of waste, while the flexural strength decreased significantly over 5% of addition. Tests with 35% of PC-TV glass, in total substitution of feldspars, highlighted a strong development of closed porosity, which interrupted the sintering process and led to bloating phenomena (Andreola et al., 2008). Even if three out of four papers followed an industrial-like cycle, there are too few data to demonstrate a technological feasibility for waste amounts higher than 5%. Glass addition to a clay body (up to 10%) to obtain vitrified tiles induced a higher firing shrinkage and improved the

bending strength. However, higher glass amounts (20%) saw a drop of shrinkage, mechanical properties, and also bulk density values (Andreola et al., 2010). PC-TV screen forms a liquid phase with a lower viscosity with respect to the SLS, which accelerates the densification kinetics but also the expansion phenomena, due to coalescence of closed pores (Andreola et al., 2010). Available data for red stoneware indicate the best technological performance with 5% of PC-TV screen glass.

*J3. PC-TV cathodic tube* is the hidden part inside the PC-TV set composed of a low melting temperature glass, enriched in Pb. For this reason, CRT has been investigated as flux in substitution of feldspars in porcelain stoneware (up to 10%) sometimes coupled with SLS up to 5% (Rambaldi et al., 2004; Tucci et al., 2006; Raimondo et al., 2007).

The few data available on batches with CRT agree on the decreasing of firing temperatures, no significant variation of firing shrinkage and water absorption, and even a slight improvement of bending strength. Nevertheless, available technological data do not allow any recommendation about the CRT amount recyclable. Although a quite complete inertization of lead into the ceramic matrix was

found through a leaching test (Raimondo et al., 2007), the ban of Pb in force in the ceramic industry constitutes an unavoidable hindrance, since market often explicitly requires leadless products.

*J4. Lamp glass* includes different types of waste glass: incandescence bulb, neon (Chemani, 2012), fluorescent lamps (Morais et al., 2011; Araújo et al., 2014; Andreola et al., 2016) and even mixed lamps (Gaitanelis et al., 2018). Their heterogeneous composition represents a challenge for recycling because the different waste lamps are generally recovered all together. In addition, these mixtures of various glasses vary randomly, according to the changing customs.

Significant technological data are given only for glasses from incandescence (Chemani, 2012) and fluorescent lamps (Morais et al., 2011) used in substitution of feldspars (up to 15%) and clays (up to 10%) in porous tiles. Since fluorescent bulbs contain a Hg lamp, a descontamination process was preventively applied (Morais et al., 2011). The technological behavior confirms what already observed for other glasses (such as SLS) in clay bodies, with the increasing of firing shrinkage and bending strength, and lowering of water absorption. The performance of porous tiles was improved for all the tested percentage of waste glass; therefore, the recommended amount is in the 10-15% range.

*J5. Other post-consumer glasses* stem from solar panels, substantially similar to SLS (Lin et al., 2015), as well as TFT-LCD (Lin, 2007; Kim et al., 2015, 2016) and veal (Gualtieri et al., 2018) that are both rich in boron and alumina. Unlike LCD produced before 2010, the current glasses do not contain toxic components, such as arsenic and antimony oxide (Kim et al., 2015). Only two papers report technological results on LCD glass in replacement of feldspars (up to 40%) in porcelain stoneware (Kim et al., 2016) or in place of clays (up to 50%) in porous tiles (Lin, 2007). Anyway, data are incomplete or unsatisfactory, so it is not possible to infer a technological feasibility of waste recycling. In

the case of porcelain stoneware, the only two properties investigated (water absorption and bulk density) indicate a firing expansion with loss of bulk density after the addition of 10% LCD glass. As concerns tests on porous tiles, the final products did not reach satisfying technological performances.

*J6. Glasses from waste melting* derive from different vitrification processes: municipal solid waste incinerator fly ashes by the application of thermal plasma melting (Bernardo et al., 2011); polluted dredging spoils from industrial area (Brusatin et al., 2005); feldspar mining residues and lime from fume abatement system of the glass industry, admixed with SLS (Bernardo et al., 2008a, 2008b) or PC-TV glass (Bernardo et al., 2008b); air pollution control residues from thermal energy plants (Deveraj et al., 2010). Overall, these wastes have a low percentage of silica but are rich in alumina and particularly CaO. Air cleaning residues have a problematically high chlorine (~2%). The glass obtained by plasma melting (Deveraj et al., 2010; Bernardo et al., 2011), as well as those from mining residues vitrified with lime and SLS (Bernardo et al., 2008a), were used in place of the non-plastic component (up to 70%) in a novel kind of “glass-ceramic stoneware”. Otherwise, the waste derived from a mixture with PC-TV glass was used in substitution of feldspars (up to 7%) in a porcelain stoneware batch (Bernardo et al., 2008b). Results regarding porcelain stoneware are discriminated upon waste amount: with a small percentage, tolerable variations of the final properties were recorded, and a 7% waste addition can be recommended (Bernardo et al., 2008b). On the other hand, the use of large quantities (>50wt%) of glass waste rich in alumina, lime and alkali induced a strong crystallization of feldspars, gehlenite and wollastonite during sintering (Deveraj et al., 2010; Bernardo et al., 2008a). Although the physical performances were good (high bulk density and flexural strength), finished products do not match standard and aesthetic requirements for commercial tiles.

Vitrified dredging spoils, being characterized by a high Fe content, were used in replacement of clay (up to 10%) in red stoneware (Brusatin et al., 2005). Firing behavior of these clay bodies confirmed what already observed for other waste glasses: technological properties can be improved (higher flexural strength and lower water absorption, even if shrinkage was greater) for addition up to 10% (Brusatin et al., 2005). The results here reported for melting wastes cannot be generalized. Since their chemical composition strongly depends on the types of residues involved in the vitrification treatment, their use has to be evaluated case by case.

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