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Incorporation of solid wastes in red ceramics – an updated review

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ABSTRACT

A review on the fundamentals and technological advances in the incorporation of industrial solid wastes into red ceramics was updated since 1997, when the first overview on this subject was published by Dondi, Marsigli and Fabbri. Other works conducted before this period, but not covered by that overview were also reviewed. Modifications were introduced in the original categories of wastes to permit a wider variety to be considered. In addition to fuel and fluxing wastes, a category of property affecting wastes substituted the originally proposed fly-ash and plasticity reducing/ plastifying wastes categories. In order to be more comprehensive, this updated review considered industrially relevant subdivisions within each category. Accordingly, fuel wastes encompass: oily residues, blast furnace sludge, and paper industry residues. Fluxing wastes encompass: rejects from ornamental rocks, glassy residues and fluxing ashes. Property affecting wastes encompass: grog, water treatment sludge, steel-refining sludge/slag, non-fluxing ashes, mineral processing tails, galvanic sludge, spent catalyst reject, textile slurry, foundry reject sands, tannery sludge and construction and demolition leftovers.

Keywords: solid wastes, incorporation, red ceramic, updated review.

1 INTRODUCTION

Today's industrial activities and human consuming habits are generating an ever growing amount of all kinds of wastes. In response, considerable efforts are being conducted to find practical and economical uses for any existing waste [1]. If viable, the waste should then become a by-product with an associate market value. For instance, PET plastic bottles and aluminum cans are now regularly collected as valuable raw materials for recyclable products [2]. In spite of these efforts, a huge quantity of innumerable types of gas, liquid and solid wastes are increasingly being disposed into the environment without treatment and generating serious consequences [3].

Gaseous and liquid wastes can be especially harmful owing to their easy dispersion in the atmosphere and water bodies [4]. As a solution, these fluid wastes should be collected and conveniently treated before being released to the environment. In particular, gas wastes represent a formidable challenge to our society mainly by their contribution to climate change [5]. Fast degradable organic wastes, associated with urban garbage, crop residues and food processing industries can also contribute to climate changes. Organics are naturally converted into secondary gas emission. The carbon dioxide and methane that result from this conversion are greenhouse gases responsible for the global warming [6]. Incineration, compostation and fuel production are nowadays proper solutions for these organic composed solid wastes, as long as resulting gas emissions are controlled [7,8].

By contrast, non-organic solid wastes, such as mining tails and industrial residues as well as civil construction and demolition leftovers, do not represent an immediate threat to the climate but are, nevertheless, affecting the environment [9]. These wastes, including low oily content petroleum residues, whether dried or as slurry, are still being improperly disposed in ways that cause long lasting pollution. Land-filling has been a commonly applied solution [10]. However, this practice can also be associated with ground water contamination [11] and deleterious changes in the landscape, mainly by invasions of ocean coast lines and river borders [12]. A technically efficient and cost-effective solution is the incorporation of solid wastes into relatively low cost and inert products that are manufactured in large amounts. Red ceramics such as bricks and roofing tiles made from clayey raw materials is currently one of the best alternatives for solid wastes incorporation [13]. Owing to the nature of the clay minerals [14,15], this incorporation also permits

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relatively small amounts of toxic wastes such as heavy metals to become safety dispersed in the ceramic body [16,17].

2 OBJECTIVE OF THE WORK AND CATEGORIES OF WASTES

Based on the above considerations, the main objective of this work is to present an updated review of the fundamental and technological advances related to the incorporation of different kinds of solid wastes into red ceramics. The work will concentrate, but will not be restricted, to investigations performed in the past decade, following the first general articles related to the subject [18-20]. In particular, Dondi et al. [19,20] published in 1997 a two parts overview on the recycling of industrial and urban wastes into clayey bodies for brick production. This overview was based on a literature survey covering a period since 1977, and included publications from a few selected countries such as Italy, UK, Spain, Germany and the USA. Already existing works from other countries were disregarded by these authors. The basis for the overview was a classification of the wastes into four categories associated with the main effects caused to the ceramic: (a) fuel, (b) fly-ash, (c) fluxing and (d) plasticity reducing and plastifying wastes. A substantial amount of information was presented by the authors in terms of the characteristics of the wastes, process parameters, and properties of the incorporated ceramics. A discussion on the complete work of Dondi et al. [19] is beyond the scope of the present work, which intends to be an updated contribution. A similar framework in terms of waste classification will be used but significant modifications are introduced to permit a wider variety of residual substances to be considered as possible red ceramic incorporated wastes.

Contrary to Dondi et al. [19], this work proposes another more general classification based not only on the specific nature of the waste but also on the processing and properties of the ceramic product. Consequently, it is here suggested to move Dondi *et al.* [19] fly-ashes and plasticity reducing and plastifying wastes to two more comprehensive new groups. In fact, as further discussed, fly-ash is a very specific kind of waste that can be included in other categories depending on its composition. Moreover, a plastic or non-plastic behavior of a waste depends basically on the presence and amount of clay minerals [14,15]. In practice, any kind of waste, being it a fuel or fluxing, without clay minerals, will behave as a plasticity reducing one. On the contrary, any waste with predominance of clay minerals and particle size below 2 μm will act as a plastifying one. It should also be mentioned that any solid waste, depending on its particle size and firing temperature, will always affect the property of the ceramic. In other words, as far as incorporation into red ceramic is concerned, there is no such a thing as an inert waste.

Accordingly, the three categories presently proposed are:

- Fuel wastes – with a carbonaceous content, which can provide a sensible energetic contribution to the ceramic sintering process;
- Fluxing wastes – with a content of alkaline and earth alkaline compounds that form liquid phases that contribute to relatively low sintering temperatures;
- Property affecting wastes – with substances that modify the ceramic behavior and cannot be included in the first two categories;

3 FACTORS AFFECTING THE INCORPORATION

It is important to observe that the first two categories proposed in the present work, fuel and fluxing wastes, normally change the properties of the fired ceramics and, in principle, they could also be included in the third category of property affecting wastes. However, due to the peculiar characteristics of these wastes, the present work agrees that they deserve to be associated with proper categories as originally proposed by Dondi et al. [19]. By contrast, although produced in large amounts by coal thermoelectric plants, ashes (fly or bottom) are just another kind of waste among hundreds of specific ones. Therefore, ashes should not be associated with a specific category.

It is also relevant to comment on the economical and environmental factors that affect solid waste incorporation into ceramics. Regarding the cost, one must realize that, in many instances, the waste producer gives it as free of charge or even pays to get rid of it. In these cases, depending on the cost of transportation, the ceramic industry is willing to accept the waste as long as it does not impair the quality of its products. However, there is a growing tendency of the producer to consider the waste as a by-product and to commercialize it at a given price. In this case, the ceramic industry will accept the incorporation only if a significant saving in energy or a considerable enhancing in the ceramic quality occurs. Environmental aspects may also restrict the incorporation of wastes. Nowadays, the majority of governmental agencies specifies the maximum levels of elements and compounds in emissions caused by firing the incorporated ceramic or by leaching/solution tests on the final product. Considering all these factors that affect the incorporation, the present work discuss results related to each proposed category of waste.

Finally, it should also be mentioned that the incorporation of a solid waste can interfere with the processing of the ceramic by imposing constraints to the molding equipment or affecting the energetic balance associated with the firing stage.

4 FUEL WASTES

This category of waste has, as its main characteristic, a relatively high heat power due to combustible carbon containing matter. A fuel waste is, in principle, desirable for its saving in energy, an always welcome condition by the ceramic industry. However, it also brings limitations associated with ceramic porosity and atmospheric emissions. In particular, the open porosity that results from the evolution of combustion gases during firing increases the water absorption, which is an important technical requirement for building construction ceramics. In Brazil, for example, a maximum water absorption of 22% is allowed for red bricks [21]. Atmospheric emissions are also specified by environmental resolution. There is no specific resolution in Brazil for atmospheric emissions from ceramic furnaces. The most useful resolution is Conama 316 [22] that limited, for example, the maximum emissions for particulate material, SO_x and NO_x to 70, 280 and 560 mg/Nm^3, respectively.

The category of fuel waste encompasses same specific industrial residues that deserve to be separately considered.

4.1 Oily residues

These are petroleum derived wastes resulting from extraction, transportation, refining operation and industrial applications. Investigations on the addition of oily residues to red ceramics have been carried out for over 20 years, but some were not mentioned by Dondi et al [19]. Okongwu [23] was among the first to report on the effects of up to 8% addition of spent lubricating oil into clay bricks fired at 950°C. No change was observed in the water absorption and the density. On the other hand, the compressive strength of the bricks showed an improvement around 1-2% of oil addition, followed by a marked decrease. Tena *et al* [24] investigated the incorporation of residues from exhausted mineral oils into ceramic pieces for civil construction. These authors concluded that fired products can be obtained with the same or even slightly better technological properties for incorporations not greater than 5%. Earlier works conducted in Brazil [25-29], and restricted to publications in local events, presented relevant results obtained from distinct oil residues. Regarding these first Brazilian works, it is worth mentioning that, for a few percent of incorporation, an increase was observed in the red ceramic strength. Moreover, practical advantages such as increase in processing speed, reduction in equipment wear, and saving in fuel consumption were also reported.

In this past decade, several publications [30-39] provided important information on the properties of red ceramics incorporated with oily residues. In particular, the increase in mechanical strength resulting from a ten percent of oily residue was indeed confirmed and an explanations of the role of hydrocarbon content on the ceramic microstructure was proposed [30,38]. Figure 1 illustrates the change in the ceramic strength with the percentage of incorporated oily residue [38]. In summary, the incorporation of oily residues into red ceramics can be associated with advantages to clay body processing, saving in fuel consumption and enhancement of the mechanical properties.

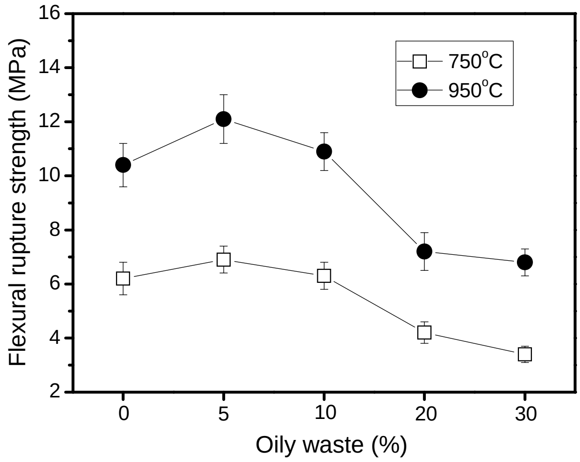

Figure 1: Effect of oily residue incorporation on the red ceramic strength [38].

4.2 Blast furnace sludge residue

The steel-making industries generate a wide variety of solid wastes [40-43], that are predominantly composed of slag, powdery materials, scraps and sludge. Most of these wastes are not easily recycled and constitute a problem for the industry. In this past decade an increasing number of papers has been dedicated to the properties of red ceramic incorporated with steel-making industry wastes [44-57].

The blast furnace sludge can be considered a fuel waste because it still has a significant amount of coke, which may reach more than 25% [44-47]. In such a case the blast furnace sludge could substantially contribute to an energy saving during the firing of the red ceramic. However, mineralogical content of blast furnace sludge could impair major required properties of the incorporated ceramic such as the mechanical strength. The blast furnace sludge is predominantly composed of Fe oxides (hematite, magnetite and wustite) as well as quartz, calcium carbonate (calcite) and coke. This sludge residue showed a fine particle size that is appropriated for incorporation into clayey ceramics. By doing so, it contributed to increase the water absorption and to decrease the mechanical strength of the fired ceramic [47], according to Fig. 2. This is due to the presence of carbon and the action of Fe compounds. The recycling of this waste into clayey ceramic can be done in low amounts, probably around 5 wt.%.

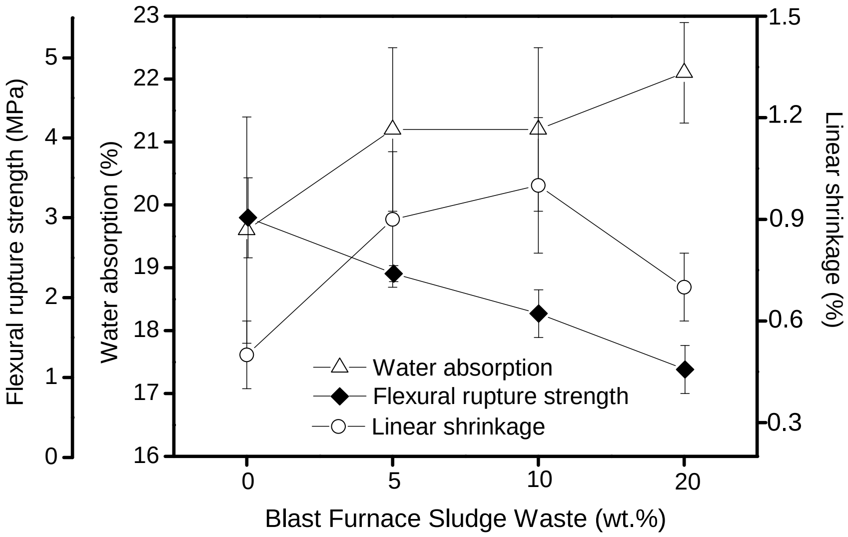

Figure 2: Fired technological properties of the elaborated compositions [47].

4.3 Paper industry residue

The pulp and paper making industry generates effluents that present serious problems related to their disposal, even after the treatment. Among the solid wastes generated by this industry, the main one is the primary sludge resulting from the effluent treatment plant (ETP). After the initial treatment, the decanted solids go through a dewatering stage generating a solid waste denominated primary sludge. This primary waste is composed basically of kaolin, wood pulp, traces of chemical substances and water. The characteristic of this waste depends on the process and the employed techniques. It can vary significantly from one industrial unit to another, even in the case that the final products are similar. Its composition is approximately 60% of kaolin and 40% of pulp, and these percentages can vary with time as a function of the process [58-60].

The primary waste from the paper industries has been successfully incorporated into red ceramics [61-65]. This is another type of waste that can also contribute to a saving in energy during the firing stage; a fact that has motivated a growing number of publications in recent years. For instance, Pinheiro [65] characterized a typical waste generated by a paper making industry and evaluated the effect of its incorporation up to 10 wt.% on the properties of a clayey body used for brick production. The results showed that, in agreement with the characteristics of the waste, its recycling into red ceramic contribute to decrease the consumption of fuel during the firing stage. However, the waste changes the physical and mechanical properties, generating a deleterious effect on the water absorption, Fig. 3, and the flexural rupture strength of the ceramic.

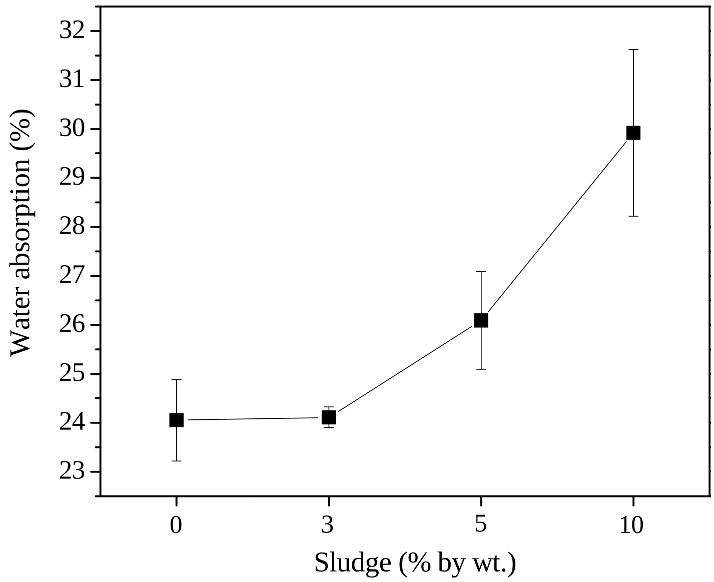

Figure 3: Water absorption of the investigated compositions firing at 750°C [65].

Environmental analysis was performed in industrial scale by monitoring the atmospheric emissions during the firing of the ceramic incorporated with 10 wt.% of the same paper making waste, using eucalyptus firewood as fuel. The evaluated parameters associated with particulate materials (PM), SO_2 and NO_x, Table 1, are in accordance with the Brazilian norms [22,66]. Consequently, the level of incorporation investigated in this work does not impair the quality of the air or causes harmful effects either to the environment or to human health.

The solution tests showed that the incorporated fired ceramic also fulfill the requirements of the Brazilian environmental standard [67].

Table 1: Values obtained for atmospheric emissions monitoring during the industrial firing of 10 wt.% waste incorporated brick as well as maximum limits allowed by CONAMA (Brazilian Environmental Council) resolutions [22,66].

Parameter	**Result (mg/Nm^3)**	**CONAMA N° 316**	**Result ($g/1 \times 10^6$ kcal)**	**CONAMA N° 008**
PM	55.7	70	100.07	350
SO_2	0.96	280	2.02	5000
NO_x	32.79	570	0.03 kg/h	–

5 FLUXING WASTES

Fluxes for ceramic processing are raw materials with a relatively high amount of alkaline oxides, mainly K_2O and Na_2O, which in reaction with silica and alumina promote liquid phase formation that facilitates the densification. The liquid phase surrounds the solid particles and, by surface tension, enables the particles to approach each other and so close the porosity, which improves the structural compactation of the ceramics [68-70].

5.1 Sludge wastes from ornamental rocks

The waste in the form of a sludge resulting from the sawing operation of ornamental rocks, such as granite, is a potential raw material for ceramic incorporation that could serve as a flux source. Its relatively low or practically no cost can be an economic advantage to the red ceramic sector. Granite is considered a flux material due to its large amount of alkaline oxides. These oxides derive from feldspars and micaceous minerals that are common constituents of granitic rocks. Related works have utilized granite waste or other kind of rock from sawing operations in the production of ceramic [71-83]. The major observations were that granite waste shows physical and mineralogical characteristics similar to the raw materials used in the body composition. Moreover, the technological properties of some mixture fulfill the required properties for the products.

Some results obtained by the incorporation of granite waste from sawing operations into red ceramic bodies for bricks, indicated a reduction in mechanical strength and increase in water absorption. For example, Vieira et al. [73] studied the effect of granite powder waste incorporation in a red ceramic body. The results indicated that granite waste presents favorable characteristics for addition into red ceramics due to facilities on the drying stages and decrease in porosity. Figure 4 shows the pore-size distribution curves for the elaborated mixtures fired at 970^oC in industrial furnace [73]. These curves demonstrate that the granite waste incorporation played an important role both on the amount of porosity and the pore-size distribution. The volume of pores decreased with increasing granite waste while the pore-size distribution becomes gradually finer. This is due to the increase in the amount of fluxes, K_2O and Na_2O, that results in a liquid phase sintering process (vitrification). The decrease in weight loss with granite waste addition also contributes to the reduction of the open porosity.

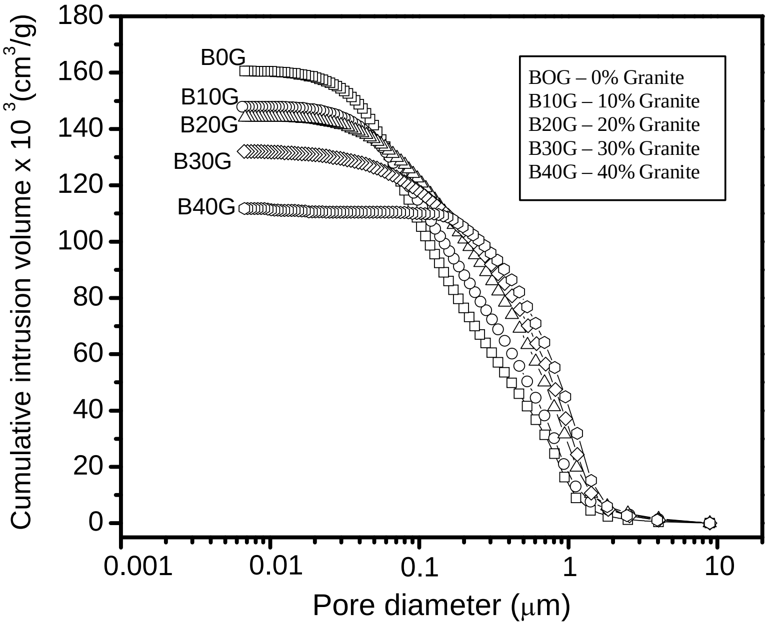

Figure 4: Porosimetry curves for the different granite compositions of clay bodies (B) fired at 970^oC [73].

5.2 Glassy residues

These glass-containing wastes correspond to all kinds of scrap products or broken pieces such as bottles, lamps, bulbs, small flasks, window plates, mirrors, glass fibers and mats. Although a common soda-lime glass residue is non-hazardous and can be easily recycled owing to their low melting and processing temperatures, a huge amount is still discarded in city dumps and landfills. The incorporation of glassy residues to clay ceramic products is a viable alternative due to the good compatibility between the clay and the common soda lime glass structures. Since more than 50 years ago, works have been dedicated to the incorporation of glass to clayey ceramics [84-92].

In 1957, Everhart [84] reported that the incorporation of more than 2.5 wt.% of glass improved the strength and water absorption of a clayey ceramic. Shutt et al. [85] developed bricks with varying glass content. Youssef *et al.* [86] mentioned that the addition of 33 wt.% of soda lima glass to a kaolin based ceramic was a promising procedure to fabricate floor tiles. Matteucci *et al.* [87] observed that the glass addition to a clay body did not significantly alter the processing and properties of all tiles. Morelli and Baldo [88] found that the introduction of a glassy residue tends to decrease the firing temperature required for clay body consolidation. Moreover, both the final ceramic strength and water absorption were improved. Bragança and Bergmann [89] replaced the traditional feldspar, as fluxing agent, by conventional bottle glass to manufacture porcelain ceramic. A reduction in the firing temperature and linear shrinkage resulted. Godinho *et al*. [90] investigated the incorporation of glassy residues into red ceramics. They indicated that, in addition to a decrease in the plasticity of the clay body, the glass improved the sintering condition of the red ceramic. Furthermore, the firing shrinkage and the rupture stress increased while the water absorption decreased with the amount of added glass. Works by Pontikes and co-authors [91,92] reported that 30 wt.% of glass in substitution of roofing tile body mixtures led to ceramics with improved water absorption and strength.

5.3 Fluxing ashes

As mentioned in the objective of the present work, depending on their compositions, fly- or bottom-ashes may contain substances that form liquid phases at relatively lower firing temperatures and thus contribute to the sintering consolidation of the ceramic structure. Works related to the incorporation of these fluxing ashes are now discussed. Skrifvars *et al*. [93] reported in 1988 on the chemical composition of a wide variety of biomass ashes obtained by the combustion of several types of grass, straws, bagasses and wood leftovers. The amount of K_2O and Na_2O in these ashes varied from 4.9 to 24.8 wt.%, indicating that they would have a potential to be used as a fluxing agent in traditional ceramics.

In a recent work, Aineto et al. [94] investigated the role of a new kind of fly-ash, obtained from the gasification of coal, as an addition to clay bodies. The authors found that the coal gasification ash is predominantly composed of a vitreous material with an K_2O and Na_2O content of 4.5 wt.%. Different additions were performed in two distinct clays that were then fired at 900^{o}C. The main conclusion of the work [94] was that the fly-ash favors the sintering process with a consequent reduction in the water absorption and an increase in the mechanical strength of the final ceramic product.

Although not directly related to red ceramics, it is worth mentioning that other works on the incorporation of ashes in traditional ceramics for vitrified white wares and tiles production [95-100] also reported satisfactory physical and mechanical properties.

5.4 Boron-containing residues

As a final example of fluxing wastes that have successfully been incorporated into red ceramics, the possibility of using boron-containing residues to improve the sintering process, through the formation of liquid phases, is now discussed.

Christogerou *et. al*. [101] investigated a boron waste (BW) produced from borax plants in Turkey. This waste containing 12.6 wt.% B_2O_3 was introduced in amount of 0, 5 and 15 wt.% in a heavy clay body mixture. Four peak temperatures of 800, 850, 900 and 950^{o}C, were applied to the dry pressed samples. The addition of BW in the clay body mixture results in the formation of increased amount of liquid phase during sintering. Shrinkage on sintering initiates at a lower temperature and the overall value was similar for all formulations investigated. In terms of physical-mechanical properties, for the 5 wt.% BW, sintering in the range of 900-950^{o}C presents comparable water absorption and bending strength, as compared to the reference, 0 wt.%, formulation. For the 15 wt.% BW, sintered below 950^{o}C, the bodies presented higher water absorption and reduced both bending strength and bulk density, mainly due to the greater amount of carbonates in BW, which contributes to the porosity formation. For sintering at 950^{o}C, an extended amount of liquid phase was formed, drastically reducing the open porosity, but also associated with the deformation of the final samples. The authors indicated [101] that a decrease in the soaking period to times lower than that of 4 h followed in their work could prevent warpage. As a general remark, their work suggested that the use of BW in heavy clay production could be considered feasible for small percentages of addition. However, for higher additions, one may need to optimize the sintering profile or to conduct a pre-calcination step for the boron-containing residue [101].

6 PROPERTY AFFECTING WASTES

Wastes that affect the ceramic properties encompass many residual substances that modify the ceramic properties and did not attend any condition associated with the previous categories. Basically, the relevant industrial residues that have been traditionally investigated are: grog, sludge from water treatment plants, steel-refining sludge and slag as well as non-fluxing ashes. Moreover, mineral processing tails, galvanic sludge, spent catalyst rejects, textile slurry, foundry sands, tannery sludge as well as construction and demolition leftovers are also included in this category as long as they do not supply heat power or contain fluxing substances.

6.1 Grog

Grog is the denomination given to the rejected pieces after the firing stage of the ceramics. The grog, from prefired products of refractory clays, is normally used as non-plastic material in the body composition for new red ceramic products and extruded floor tiles [102-112]. Since the grog acts as a non-plastic material, its amount and mean particle size must be optimized, according to the final characteristics desired, owing to the possibility of increasing the porosity. Whenever the grog is fired a second time above its original processing temperature, reactions and transformation may still occur.

In particular, Vieira *et al.* [107] investigated the effect of an addition of a grog fired at low temperature, not higher than 600^oC, on the technological properties of a plastic clay from Campos dos Goytacazes, typically used for roofing tile fired at 970^oC. The results showed that grog addition reduces the total linear shrinkage, Fig. 5, but does not effectively alter water absorption and flexural strength, Fig. 6. These indicate the feasibility of using grog from fired brick waste on roofing tile production.

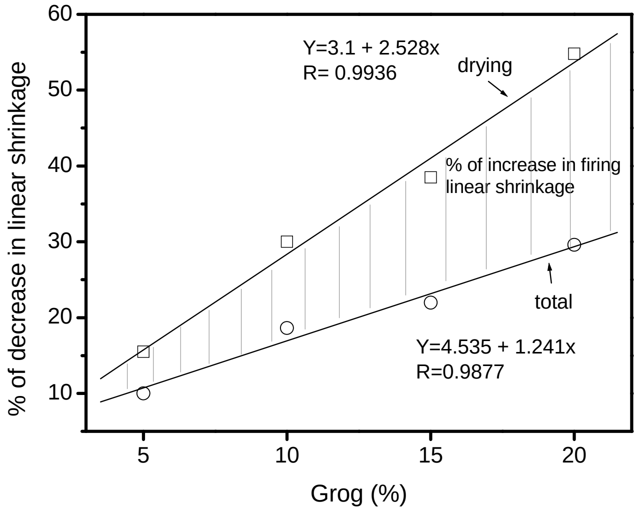

Figure 5: Correlations between the grog addition and linear shrinkage [107].

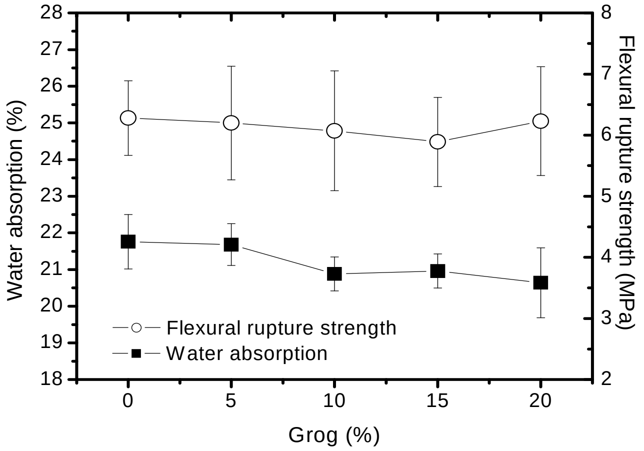

Figure 6: Water absorption and flexural rupture strength as a function of the percentage of grog addition. [107].

6.2 Sludge from water treatment plant

The treatment of water for human consumption normally generates a great amount of wastes in the form of sludges that are generally discharged in hydric resources [113]. These wastes come mainly from the decantation and filtering stages in a conventional water treatment plant. The simple discharge in a hydric resource, although a less expensive method of final disposal, is not a proper solution due to the possibility of undesirable formation of mud deposits as well as contamination from the chemical products used in the treatment. Other alternatives for the final disposal of sludge wastes from water treatment plants are presently being considered. For instance, discharge in regular sewage pipelines, accumulation in mud containing reservoirs or directly onto the soil, as landfill, are becoming common practice [113]. In all these cases, however, the waste and associated chemical products remain unaltered. An environmentally correct solution, which is nowadays being investigated [114-121], is the incorporation of these sludge wastes in clayey bodies.

As an example, Monteiro *et al.* [121] investigated the influence of the firing temperature on the technological properties of red ceramics made of a kaolinitic clay incorporated with a sludge from water treatment plant. Mixtures were prepared with amounts of 0, 3, 5 and 10 wt% of sludge incorporated into clayey bodies to fire at 700, 900 and 1100°C. Ceramic properties related to the bulk density, linear shrinkage, water absorption and flexural rupture strength were determined. The results indicated that the incorporation of the sludge increases the water absorption and reduces the mechanical strength of the clayey ceramic, as shown Fig. 7. This is a consequence of the changes caused in the porosity by the relatively elevated weight loss during the firing stage.

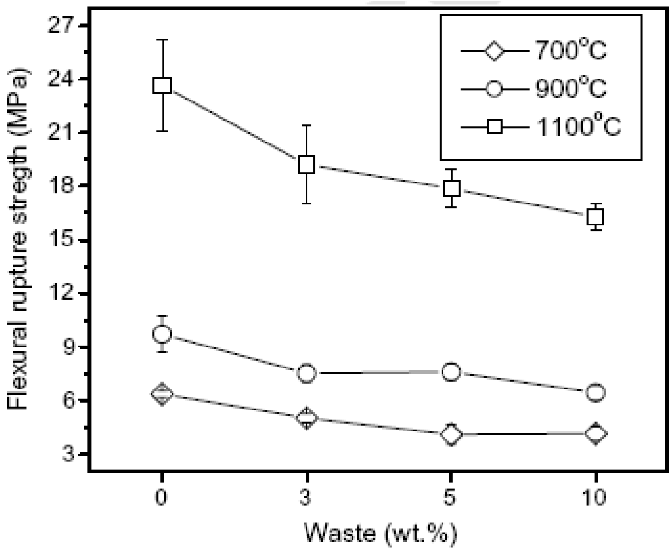

Figure 7: Flexural rupture strength of the clayey ceramic as a function the amount of decantation sludge waste addition and the firing temperature [121].

6.3 Steel-refining sludge and slag

The sludge and slag from basic oxygen steel-refining process can be incorporated into red ceramics [50-57]. However, different than the blast furnace sludge [44-47], these residues do not have enough heat power to be considered as fuel wastes. Therefore, here they are classified as property affecting wastes. Actually, these slag and sludge are another type of steel-making industry waste. But, contrary to the blast furnace sludge, their loss on ignition is practically zero, without any carbonaceous substances. It was, in general, found that the incorporation of steel-refining sludge (SRSludge) tends to increase the refractoriness of the clay and decrease the weight loss and thus reduce the ceramic porosity [52]. Up to 20% of SRSludge incorporation, there was no change in the water absorption. However, above 5% of SRSludge the strength suffers a marked decrease as illustrated in Fig. 8.

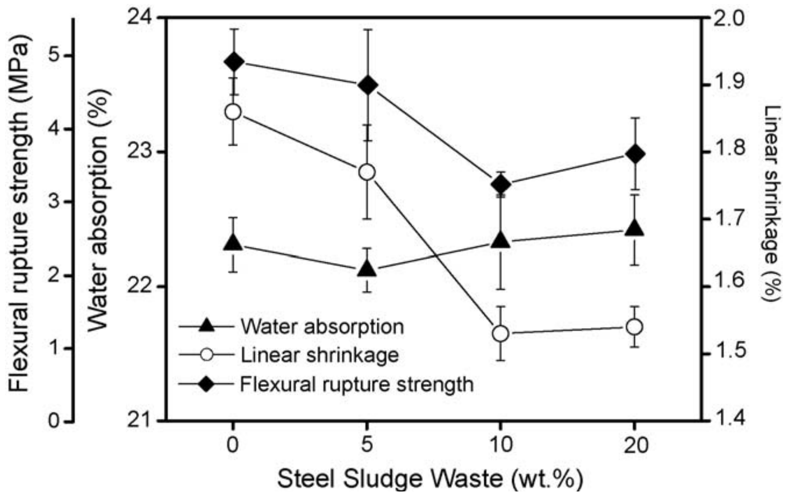

Figure 8: Technological properties of a red ceramic incorporated with steel-refining sludge [52].

It was also found that the incorporation of SRSludge into red ceramic fulfills the Brazilian standards for the extracts obtained from solution and leaching tests and, therefore, brings no harm to the environment, as shown in Table 2.

Table 2: Potentially toxic metals in the solution and leaching extracts of a ceramic with 5% steel-refining sludge [53].

Element	solution (mg/L)	Limits (mg/L)	Leaching (mg/L)	Limits (mg/L)
Al	1.8	0.2	19	-
Ba	0.06	0.7	0.7	7.0
Cd	< 0.003	0.005	0.04	0.5
Cr (total)	< 0.02	0.05	< 0.05	5.0
Cu	0.004	2.0	0.13	-
Fe	0.02	0.3	0.22	-
Mn	0.02	0,1	0.9	-
Na	10	200	ND	-
Pb	< 0.06	0.1	0.14	1.0
Zn	0.02	5.0	1.3	-

In summary, the incorporation of up to 5% of SRSludge is beneficial to the ceramic since it decrease the linear shrinkage and does not affect both, the water absorption and the mechanical strength. The incorporation can also retain potentially toxic metals in the ceramic structure without causing environmental problems.

Steel-refining slag (SRSlag) in another steel-making industry residue that can be classified as a property affecting waste. Vieira *et al.* [56] characterized a SRSlag generated from an integrated steel plant, to study the possibility of its recycling and incorporation into red ceramics fabrication. The results indicate that the SRSlag presents high amount of Ca, Fe, Mg and Si. The weight loss of the steel slag at temperatures above 800^{o}C is associated with calcium carbonate decomposition. The coarse particle size of the SRSlag was found inadequate for its direct recycling into red ceramic fabrication. As a possible solution, it was suggested that screening or grinding the SRSlag to a convenient particle size distribution would allow its successful industrial incorporation into red ceramics products

6.4 Non-fluxing ashes

A large amount of ashes produced in industrial plants has only a negligible content of alkaline compounds and thus will not act as a fluxing agent when added to clay bodies. These non-fluxing ashes, however, always affect the properties of the final red ceramic product.

The incorporation of non-fluxing ashes into clayey ceramic materials has been widely investigated [122-132]. In particular, the fly ash coming from coal-fed thermal power plants is one of the most studied. Due to the extreme variability of its physical-chemical characteristics, the incorporation of this type of fly ash in the ceramic process may result in a widely variable quality for the ceramic product. Illustrations of the effect caused by non-fluxing ashes are now presented.

Borlini *et al.* [128] investigated the use of biomass ashes, from sugar cane bagasse burnt in boilers, by incorporation into red ceramic body. The investigation was performed by evaluating the degree of imobilization of potentially toxic metals and the technological properties of fired specimens with incorporation of ash. Clay mixtures with 0, 5, 10, 15 e 20 wt.% of ashes were fired at 970°C. The results showed that these ashes are predominantly composed by quartz. The degree of immobilization of potentially toxic metals was considered satisfactory according to the Brazilian norms. However, the ash incorporation caused a detrimental effect on the mechanical strength, according to Fig. 9, which was attributed to the large particle size of the quartz.

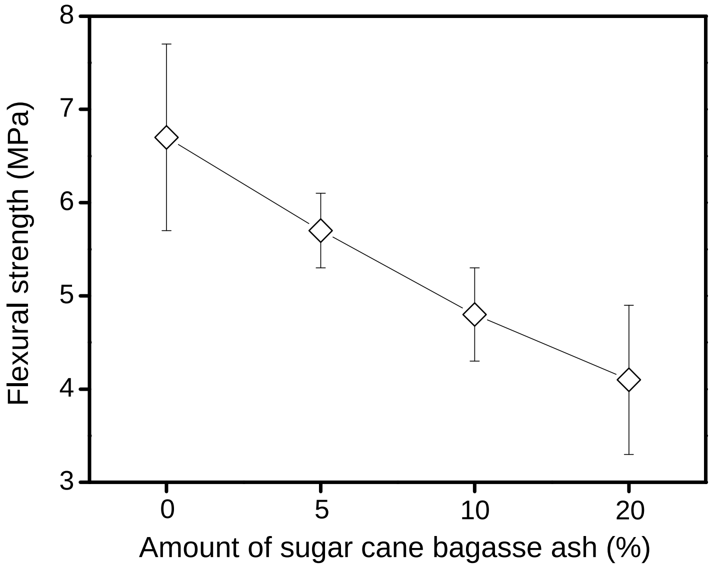

Figure 9: Flexural strength as a function of the amount of sugar cane bagasse ash added to red ceramics [128].

In another work, Borlini *et al*. [132] evaluated the influence of the firing temperature in the properties of incorporate red ceramic with sugar cane bagasse ash. Incorporations of 0, 10 and 20 wt.% of ash with granulometry inferior to 44 μm (325 mesh) were made in the industrial ceramic body. Figure 10 illustrates the diametrical compression as a function of the sugar cane bagasse ash. For all investigated temperatures, a decrease in the compression strength was observed with the amount of incorporated ash. Figures 11 and 12 show the X-ray diffraction patterns of ceramics incorporated with 0% (**A0C), 10%** (**A10C325**) and 20% (**A20C325**) of the non-fluxing bagasse ashes, then fired separately at 1050 and 1200^{o}C. The identified phases in these figures display a predominant participation of quartz (Q). In Fig. 11 and 12 it is observed that the incorporation of ash did not change the main crystalline phases. At 1050^{o}C, however, an Al and Ca silicate phase was formed.

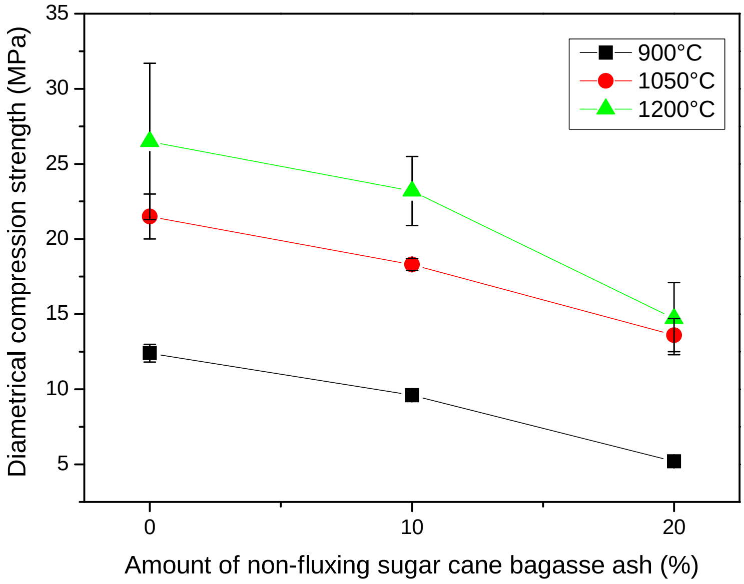

Figure 10: Diametrical compression strength as a function of the amount of incorporated non-fluxing sugar cane bagasse ash for different firing temperatures [132].

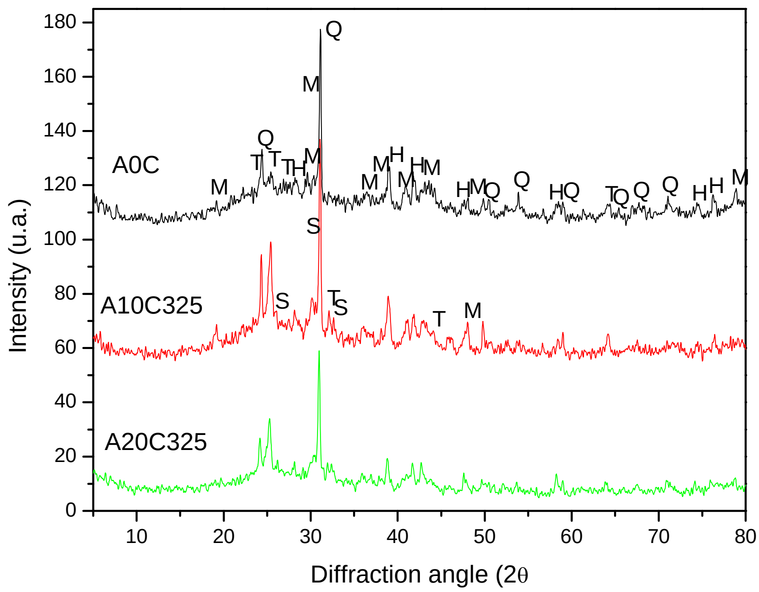

Figure 11: X-ray diffraction patterns for different compositions of non-fluxing bagasse ash incorporation into red ceramic fired at 1050°C. Abreviations: M = mullite; T = tridimite; Q = quartz (SO_2), H = hematite, S = Al-Ca silicate [132].

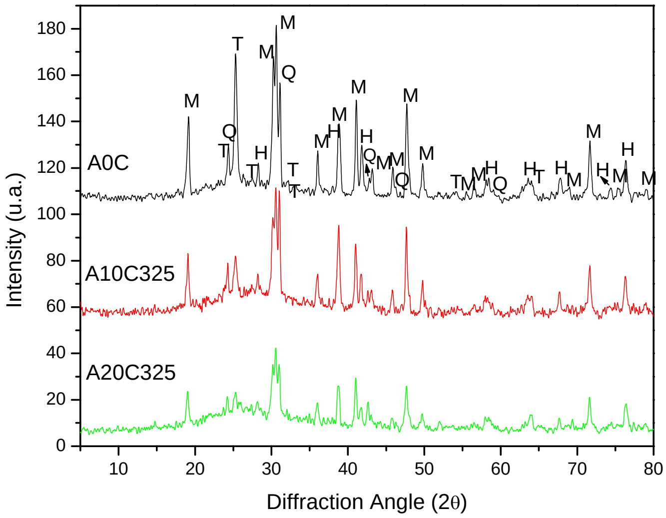

Figure 12: X-ray diffraction patterns for different compositions of non-fluxing bagasse ash incorporation into red ceramic fired at 1200°C. Abreviations: M = mullite; T = tridimite; Q = quartz (SO_2), H = hematite, S = Al-Ca silicate [132].

Figures 13 and 14 present the surface micrographs of a 1050°C fired red ceramic without ash (**A0C**) as well as incorporated with 20 wt.% of non-fluxing sugar cane bagasse ash (**A20C325**). In these figures both fractures are rough and porous. This is expected in kaolinite clay ceramics fired at 1050°C. At this sintering temperature the amount of liquid phase and corresponding viscosity are not enough to promote the total closure of open pores.

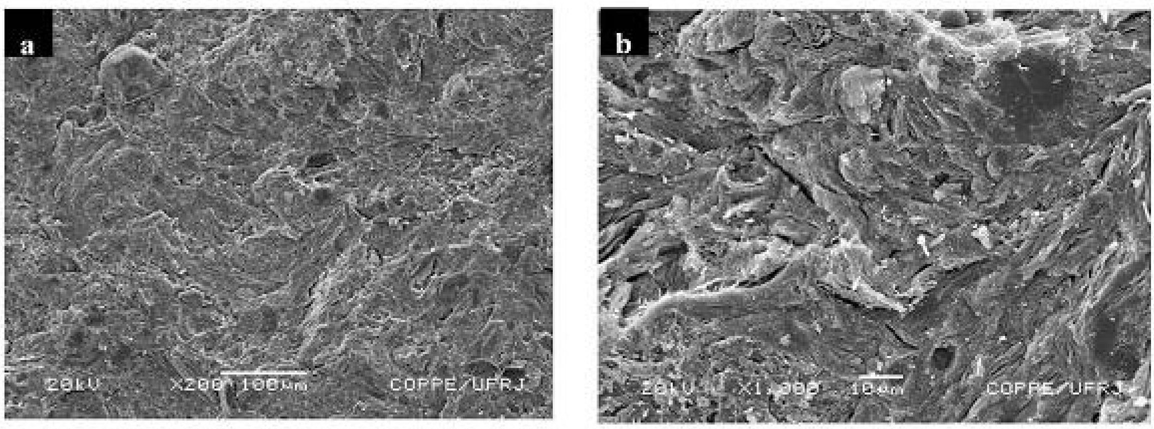

Figure 13: SEM Fractographs of the red ceramic without ash incorporation (**A0C**) fired at 1050°C. (a) 200x; (b) 1000x [132].

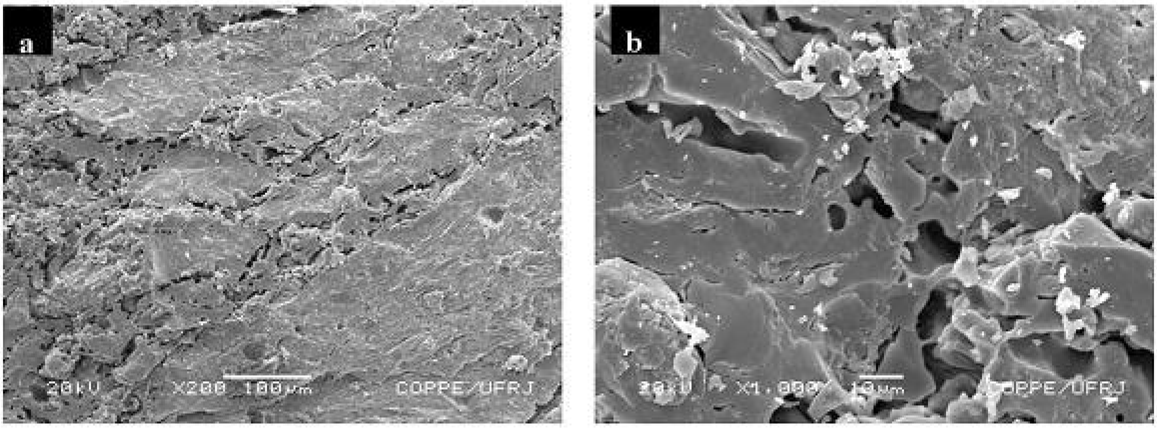

Figure 14: SEM Fractographs of the red ceramic without ash incorporation (**A20C325**) fired at 1050°C. (a) 200x; (b) 1000x [132].

6.5 Mineral processing tail

The tails or non-valuables wastes from mineral processing have been traditionally disposed in piles or sedimentations dams that represent potential impact to the environment. The possibility of incorporation of these tails to red ceramics represents a first step towards a definite solution.

In this respect, Sikalidis and Zaspalis [133] evaluated the utilization of Mn–Fe solid tails, originating from electrolytic manganese oxide production plants, as raw materials in the manufacturing process and on the properties of traditional ceramic building products such as bricks, roof or floor tiles. Ceramic test specimens incorporating 2.5, 5, 7.5 and 10 wt.% solid wastes were fired to different peak temperatures ranging from 950 to 1100^{o}C. The results indicated that the additions of these tails in clay ceramics between 5 and 7.5 wt.% improved the basic properties of the products such as water absorption and bending strength. Firing shrinkage, although slightly increased, remains always within acceptable and controlled limits. The color of the products varied from light to dark brown as a function of the added percentage of the wastes. Pilot batch experiments revealed that extruded and powder pressed products displayed properties in very good agreement with the laboratory results. As mentioned by the authors [133], these results encourage the utilization of Mn–Fe tails originating from the electrolytic production of MnO_2 in the fabrication of building ceramics. Moreover, this utilization can drastically decrease the environmental problem that is caused by the disposal of these tails.

6.6 Sludge from galvanic process

Inertization of sludges from galvanic processes into clay-based ceramics has also been investigated [134-138]. In particular, Magalhães *et al.* [134] evaluated the effect of several processing parameters such as the mixing time, the calcination temperature and its duration as well as the relative amount of sludge, and the physical aspect of the sample on the fixing level of relevant species (SiO_2, $SO4^{2-}$, Zn, Ni, Ca, Cu, Cr) by leaching in different media (aqueous, acetate, and citrate). The relative amount of sludge in the mixture, the calcination temperature and the agglomeration state of the sample were found to be the most influential parameters of the inertization process. The incipient reaction between sludge and ceramic matrix components points out for the dominance of a macro-encapsulation mechanism. The authors suggested [134] that the incorporation in a clay-based fired matrix is a promising way to immobilize different metals belonging to galvanic sludges. The firing temperature and agglomeration degree imposed by pressing of starting mixtures are the most effective experimental parameters in the inertization process. The relative amount of sludge in the mixture also plays an important role.

6.7 Spent catalyst reject

Thermal cracking units in the petrochemical industries are generating an increasing amount of spent catalyst rejects. This is becoming a worrisome factor regarding the environmental preservation. An investigation has already proposed a solution for this kind of reject.

Acchar *et al.* [139] studied the incorporation of a spent catalyst reject (SCR) in an amount of 20 wt.% into clay-based materials. The reject added clay bodies were fired at temperatures varying from 700 to

1150°C. It was found that the SCR is mainly constituted of quartz, together with kaolinite and mullite. The results indicated that, although sintering might be somewhat delayed, no major changes in the final properties of the bricks were observed. This is due to the high alumina content and the absence of high amounts of alkali ions in the reject material. According to the authors [139], the large use of the SCR into a clay-based material will reduce transportation and landfill costs as well as the need of waste disposal and the consumption of natural ceramic raw materials.

6.8 Slurry from textile industry

Textile is associated with another industrial area that has been expanding and, consequently, generating a growing amount of wastes. One work deserves attention as a possible suggestion for the solution concerning the main waste in this industry.

Herek *et al.* [140] investigated the use of 10 wt.% of a slurry from industrial laundry generated in the process of primary treatment of effluent, with the use of decantation, into clay-based material for bricks fired at 800°C. The results indicated that the tests of compression strength, water absorption and leaching and solution analysis showed values that attend the norms for bricks. In this amount of incorporation and using the techniques described in the work, it permits the use of bricks as a construction material. Additionally, this incorporation could also have the finality of decrease the environmental impact caused by the waste from the treatment of textile effluent.

6.9 Reject sand from smelting metallurgical process

Traditional foundries still use sand as the basic molding material. Even though most of this sand is recycled, a percentage is being disposed in the environment. Preliminary research works proposed the incorporation of rejected sands into clay ceramic products.

Martín-Cortés *et al.* [141] studied the use mixture of sands from smelting metallurgical process with common clay used in the fabrication of structural ceramic products such as bricks, blocks and roof tiles. Seven hundred hollow blocks containing 1 wt.% of waste were fired at 950°C. The results indicated that the incorporated bricks present sufficient compressive strength resistance for civil construction, which is the same of the traditionally bricks produced with only red clays. Furthermore, its production cost will be less than that of the traditional bricks, since less clay is used in its production.

6.10 Tannery sludge

The leather industry is considered one of the unhealthiest, both for the stinking odor and its organics pollutants. In particular, the slurry residue resulting from the treatment of the organic rejects, known as tannery sludge, is a threat to the environment. A study is beginning to indicate technical possibilities for this sludge as an addition to red clays.

Basegio *et al.* [142] reported on the results of a feasibility work on the immobilization of tannery sludge by incorporation into a ceramic product. The raw materials, tannery sludge and clay, were mixed together in different proportions. The ceramic specimens were characterized with respect to water absorption, porosity, linear shrinkage and transverse rupture strength. Leaching tests were done on ceramic bodies made with different additions of sludge. In order to evaluate the possibility of air contamination during the firing process, preliminary studies of air emissions were carried out. The results indicated that the mechanical properties of the evaluated samples were similar to those specified for ceramic bricks. All the leaching tests have shown that the main sludge contaminant i.e. chromium, could be immobilized within a finished ceramic product. The studies of air emissions have shown that zinc and chlorine are mainly collected from gas emissions and hence are not immobilized by the ceramic system. Of practical importance, the study shows that the properties of the ceramic materials produced are acceptable for applications such as bricks for the building industry.

6.11 Construction and demolition leftover

Civil construction is one of the human activities responsible for a large amount of waste generated in metropolitan areas. In fact, construction and demolition leftovers (C&DW) from city buildings may correspond to more than 30% of the total initially applied materials. Nowadays, efforts have been conducted towards solutions that avoid the accumulation of these leftovers in the environment, particularly by incorporating them into red ceramics [143,144].

The most recent work [144], investigated the addition of C&DW collected directly from building constructions in the city of Natal, Brazil, on the properties of clay-based ceramics. The results indicated that a high content, of approximately 50 wt.%, of C&DW can be incorporated into traditional red ceramic products, such as brick and tiles, without requiring changes in the processing routine or causing detrimental effects on the final product properties.

7 FINAL REMARKS

In addition to the main objective of this work, which was to present an updated review on fundamentals and technological papers related to the incorporation of solid wastes into red ceramics, it was also reviewed a relevant growing tendency of new research works throughout the past twelve years. In fact, the increasing concern of our society with respect to the rapid degradation of the environment by pollution and other practices associated with climate changes, motivated waste recycling actions not only to avoid accumulation in landfills and water bodies but also to save raw materials and processing energy.

The recycling of solid wastes by incorporation into traditional red ceramic products fulfills the intrinsic requirement of these actions. By one side, the amount of red ceramics produced all over the world is high enough to permit most wastes to be incorporated in percentages low enough not to impair the ceramic properties or bring any risk to the environment. On the other side, the incorporation of fuel or fluxing wastes contributes to a saving in energy during the ceramic firing process stage and could also significantly improve the properties of the final red ceramic product.

The present work emphasizes how these advantages are counting on the recent growth of articles related to waste incorporation into red ceramics, especially in countries such as Brazil, Portugal and Turkey, not covered by the first review papers [19,20] dealing with the subject. Furthermore, the present updated review introduces an important category of property-affecting wastes that allowed for the inclusion of different types of residual materials such as C&DW, reject sands from foundries, textile slurry and tannery sludge that only recently have been incorporated into red ceramics.

Consequently, more than just simply providing a list of works conducted in the past twelve years, this updated review discusses the importance of considering new categories of wastes and the need to pay attention to research efforts being conducted in emerging countries.

8 CONCLUSIONS

An expressive number of papers has been dedicated to the incorporation of solid industrial wastes into ceramics in the past decade, after the first overview presented by Dondi *et al.* [19] in 1997. The present work reviewed not only the decade recently published papers but also earlier papers not covered by that overview. An improved classification, in which a new category of property affecting wastes replaced the original fly-ash and plasticity reducing/plastifying wastes, was proposed. Within each category, this work introduced subdivisions related to both the nature of the waste as well as the processing and properties of the incorporated ceramic.

<u>Fuel wastes</u> with heat power, which contribute to saving in the ceramic firing energy, included: *oily residues* from petroleum operations; *blast furnace sludge* with a coke content from steel making; and *paper industry residues* with a cellulose content.

<u>Fluxing wastes</u> that promote liquid phase formation and improved sintering at lower temperature, included: *rejects from ornamental rocks* with alkaline oxides; *glassy residues* that form a low melting temperature amorphous phase; and *fluxing ashes* with enough alkaline oxides for structural consolidation by fluid phase.

<u>Property affecting wastes</u> that cover all other industrial residues not considered by the previous two categories. In this work, the following subdivision was included: *grog*, a non-plastic material formed by discarded ceramic pieces after the firing stage; *sludge from decantation and filtering* stages in water treatment plants; *steel-refining sludge and slag* with an iron oxide content in addition to other oxides; *non-fluxing ashes* resulting from combustion and incineration processes with negligible amounts of alkaline compounds; *mineral processing tail* originated from manganese oxide production plant; *galvanic sludge* from electroplating and electrorefining plants with a relatively high amount of heavy metals; *spent catalyst reject* accumulated from petrochemical cracking units; *textile slurry* from dying and cleaning stages; *foundry sand* rejected from smelting metallurgical processes that contains noticeable amounts of metals; *tannery sludge* from chemical operation with chromium salts used in leather production; *construction and demolition leftovers* corresponding to one of the largest kind of waste generated in metropolitan areas.

All papers reviewed showed a clear effort towards solving the problem of industrial waste accumulation in the environment. Furthermore, in many cases, the incorporation of the waste into red ceramics has proved to be both of economical and technological advantages.

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