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Review article

Recycling of industrial wastes in ceramic manufacturing: State of art and glass case studies

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ABSTRACT

Nowadays, ceramic tile are manufactured at zero emissions permitting to recycle all by-products and part of residues derived from depuration treatments (exhausted lime, glazing sludge and polishing sludge). In addition to this environmentally friendly tendency, in the last years an increasing number of scientific studies demonstrated the feasibility to use alternative raw materials in substitution of different component of the ternary clay-feldspar-quartz system. In the first part of the paper is reported the state of the art of industrial waste recycling in the ceramic sector, with the focus on review studies related to both ceramic tiles and bricks..

In the second part of the work are reported two case studies conducted by the authors with the aim to formulate ceramic bodies using alternative raw materials. New tailored compositions were obtained replacing clays, flux and/or inert compounds (higher than 60 wt%) by scraps from packaging waste glass in tiles, and cathode ray tube glasses and packaging waste glass up to 20 wt% in the brick compositions.

Contents

1. State of the art of industrial waste recycling in ceramic sector

The amount of inorganic wastes (derived from Construction & Demolition and Mining & quarrying activities) in Europe is estimated to be more than 1,500 million tonnes [1]. The continuous increase of waste amount requires not only measures that reduce its generation, but also recycling and recovery. In this regard, the European directives concerning waste, Directive 2006/12/CE and Directive 2008/98/CE, are oriented to transforming the European Union into a "recycling society" that attempts to avoid generating waste and that uses its waste as a resource [2,3]. Traditionally, non hazardous inorganic wastes have been disposed off in landfills and often dumped directly into ecosystems without adequate treatment but possible recovery or recycling alternatives should be investigated and implemented [4].

On the other hand, the world production of bricks and ceramic tiles requires massive amount of natural raw materials, since, until now, it is based mainly on the traditional clay-silica-feldspar system.

Natural raw materials used in the fabrication of clay-based ceramic products show a wide range of compositional variations and the resulting products are very heterogeneous. Therefore, such products can tolerate further compositional fluctuations and raw

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material changes, allowing different types of wastes to be incorporated into the internal structure of ceramic tiles and bricks as part of their own matrix.

Regarding ceramic tiles processing, several studies made in the last decades are related to the substitution of conventional raw materials by other natural resources as zeolites [5,6], volcanic rocks [7,8] and nepheline syenite [9]. Besides, the quartz substitution by Si-rich waste as rice husk ash and silica fume was studied in whiteware compositions with good results concerning the reduction of sintering temperature (~50 °C) [10].

Other works have investigated the use of different alternative fluxing agents (soda–lime glass cullet [11], cathode ray tube of TV or PC monitor (CRT glass) [12,13] and granite cutting sludge [14]) substituting partially the feldspars to obtain sintering behaviour and mechanical characteristics similar to the industrial compositions. It is well known that the excessive addition of fluxes leads to an increase of amorphous phase in the final product with a negative effect on the mechanical properties. Research on the use CRT glass as fluxing agent highlighted that 5 wt% is the optimum amount of glass that it is possible to replace for Na-feldspar [12]. A complete substitution of Na-feldspar by glass (35–40 wt%) is not feasible because of the relatively low viscosity of parent glass (soda lime or CRT glass) at firing temperatures, particularly enriched in alkali and alkali-earth oxides [11–13]. According to these studies [12,13], the composition containing CRT glass starts the sintering process already at 850 °C, temperature lower than the traditional one, and at 1150 °C the open porosity is practically eliminated. The formation of closed porosity, however, starts at 1100 °C when open porosity still has a too high value of 14%. Then closed porosity increases with the temperature reaching 18% at 1200 °C; as a result, the total porosity remains always very high. However, due to the intensive formation of closed pores, the shrinkage rate significantly decreases, the final shrinkage is too low (3.5%) and at 1200 °C the deformation without a complete densification begins. The high amount of alkali glass in substitution of Na-feldspar in porcelain stoneware bodies provokes an unusual densification behaviour which leads to a not fully densified product [11–13].

More recent works have studied alternative fluxes characterized by a high crystallization trend, in this way the crystallinity of final ceramics is increased. This effect can be obtained using glass-ceramic frits [15] or waste residues with high crystallization tendency (as slag and bottom ashes) [16,17]. In this case, due to recrystallization processes during the sintering and cooling steps, the amount of residual amorphous phase decreases, leading to an improvement in the mechanical properties [15].

Other possibilities to use non traditional raw materials within the ceramic body formulation is offered by ceramic tiles with insulation properties. With the increasing emphasis on environmental protection and energy conservation, these ceramic products have now a considerable market as building materials. These tiles have a porous body, with a bulk density of about 0.6–1.0 g cm^{-3}, while the surface is glazed so both insulation energy-saving effect of porous material and also the surface cleanability of glazed tiles are ensured. For these applications several compounds with pore forming properties can be use: inorganic residues containing calcium carbonate, as egg shell or glass cutting sludge, or wastes with organic nature, as agro waste or char pyrolysis [18–20].

With the aim to formulate ceramic bodies using high amounts of alternative raw materials, new tailored compositions can be obtained replacing clays, flux and/or inert compounds (> 60 wt%) using scraps from packaging glass waste [21–23].

Regarding brick-making, numerous researches are related to the use of different kinds of wastes in brick body. Heavy clay ceramics, as bricks, are produced from natural raw materials with a very wide–ranging overall chemical and mineralogical composition. For this reason, such materials can tolerate the presence of different types of urban and industrial wastes (fly ash from coal-burning thermal power plants, sludge from the glazing lines of ceramic tile manufactures, sludge from plating plants of metallurgical and mechanical industry and from the mining industry), even in considerable percentages [24,25]. This is a strategic action by considering that in recent decades, the growing consumption and the consequent increasing of industrial production has led to a fast decrease of the available natural resources (raw materials or energy sources). Therefore, alternative ways to reuse several types of waste materials have been attempted in recent years, including the incorporation in clay brick products.

Depending on their principal effect, the wastes used can be divided in three main categories: (i) fuel wastes, (ii) fluxing wastes, and (iii) plastifying wastes.

Fuel wastes are generated from a wide range of industrial production processes: residues of the urban wastes treatment plants, extraction and carbon manufacture, textile and tanning industry, oil, paper, agricultural and wood industry [24–34]. Thanks to their organic substances content, during their combustion, they bring an energetic support in the bricks firing phase and contributes to increase their porosity when added into the body. Usually, the addition of this kind of waste is limited to 10 wt% in order to reach an equilibrium between positive and negative. In the developing countries of Africa and Asia, the use of agricultural residues instead of some part of the expensive primary fuel, include rice husk, sawdust, straw, maize cobs, and animal dung is a widespread practice.

In recent years, the new challenges of environmental protection and energy saving have provided building materials made with non-traditional raw materials, having thermal insulation properties and requiring low energy-consumption: these ceramic bricks should have a considerable market. In this context the use of agro-waste as pore forming agent is a viable route to increase porosity. Sawdust and grapes and cherries seeds, thanks to their organic substances content, during their combustion, bring an energetic support in the bricks firing phase and act as pore forming agent. The addition of this kind of waste was limited to 5 wt% in order to reach an equilibrium between positive (weight and shrinkage decrease and porosity increase) and negative (increase of water absorption and mechanical resistance decrease) effects. [33]. The addition of sawdust provokes an important reduction of samples weight (~20%) due to the presence of porosity but the mechanical properties are lower than the industrial tolerance values. Regarding grapes and cherries seeds, the addition of 5 wt% to a brick formulation showed better results with respect to the sawdust, maintaining the mechanical properties of the fired brick (950 °C), showing flexural strength values around 21–22 MPa higher than the industrial tolerance with a weight reduction of 3–10% [33].

Fluxing waste, as sludge from the glazing lines of ceramic tiles manufacture and from plating plants of metallurgical and mechanical industry, are added in percentage lower than 20 wt% to reduce the final porosity of the bricks, and to improve the workability in the extrusion phase, but they can cause a decrease (15–40%) in the mechanical resistance of the dried products [35,36].

Plastifying waste such as residues from mineral and metallurgical industry are capable to express a filler and plasticity modifiers role. Many materials, thanks to their coarse particle size distribution, are used in order to control the plasticity typical of the clayey raw materials and to obtain a useful level of workability with a reduced water consumption. The amount of waste added to the body can varies from 10 to 60 wt% and causes a decrease of the shrinkage and the mechanical resistance of the fired bricks. Normally, residues of stone treatments as granite or marble cutting