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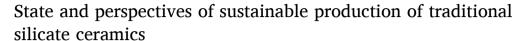
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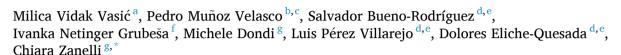
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Perspective





- ^a Institute for Testing of Materials IMS, Bulevar vojvode Mišića 43, 11000, Belgrade, Serbia
- ^b ESIT. Universidad Internacional de La Rioja, Avda. de la Paz 137, 26007, Logroño, Spain
- ^c Facultad de Ingeniería, Universidad Autónoma de Chile, 5 Pte, 1760, Talca, Chile
- d Department of Chemical, Environmental, and Materials Engineering, Higher Polytechnic School of Jaén, University of Jaen, Campus Las Lagunillas s/n, 23071, Jaén, Spain
- e Center for Advanced Studies in Earth Sciences, Energy and Environment (CEACTEMA), University of Jaén, Campus Las Lagunillas s/n, 23071, Jaén, Spain
- ^f Department for Civil Engineering, University North, Ulica 104. brigade 3, 42000, Varaždin, Croatia
- ⁸ CNR-ISSMC, Institute of Science, Technology and Sustainability for Ceramics, 48018, Faenza, Italy

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ABSTRACT

The traditional ceramics industry uses large amounts of primary mineral raw materials. Improvements in the production of building materials based on non-metallic minerals can contribute to sustainable development in many ways, such as saving natural resources, using waste materials, reducing energy consumption, decreasing emissions hazardous to the health and the environment, particularly carbon dioxide, and reclamation of mines after exploitation of raw materials, etc. This paper describes the state of application of mineral raw materials and waste in the traditional ceramics industry with a perspective on future challenges. Intensified research is needed to complement the laboratory data and re-scale to the industrial-sized products while improving communication between both sectors.

1. Introduction

Roadmaps urged by the European Commission for the ceramic industry lay out ambitious long-term objectives for resource efficiency, energy savings and a low-carbon economy. Initially set for 2050 as their target year [1,2], they are recently anticipated to 2030 and include halfway goals [3].

Building materials of non-metallic origin represent, despite great technological development, still the essential materials in construction. Also, despite the series of difficulties it faces today, this industrial sector is one of the most vital in many countries. After processing of the ceramic materials, after the discard in production, or at the end of life, these can be re-used in modern technological conditions, which gives them the potential of recyclable resources. Although the production rate was highly reduced due to the financial crisis of 2007–2008, since the last decade, the manufacturing of construction and building materials

has been slowly but steadily rebounding. For instance, in Spain, which is one of the largest producer of fired bricks in the EU, the production rate has increased by more than 60 % since 2014 [4].

Natural raw materials are the basis for the production of building materials and belong to non-renewable natural resources, given the time needed for their creation and the limited amounts. The continuous increase in the growth of industrial production imposed a multiplication in the amount of waste materials from various production processes. Some of the industrial waste has been reused today, and they represent secondary raw materials of technogenic and anthropogenic origin. The degree of recycling, however, is extremely variable in time and space, depending on economic, technological and social conditions, and the legal framework as well. The initiative to create standards for secondary raw materials quality and a developed market is necessary for the ceramics industry [5]. Besides, the market reacts to "green products" in a strongly differentiated way [6], often varying from country to country.

E-mail address: chiara.zanelli@issmc.cnr.it (C. Zanelli).

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^{*} Corresponding author.

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There is a general need for a better flow of information about environmentally-friendly products to consumers [6-8].

In the traditional ceramics industry, the key factors are the high-level consumption of natural resources and energy (most often from nonrenewable sources), flue gases and large amounts of waste after the useful life of the product [8]. Another problem is the need for rehabilitation and reclamation of mines after extraction. Given that the ceramic industry can consume a large share of waste materials due to a wide range of fluctuations in their composition, the research focus of recent decades has been mostly directed toward this subject. Bearing in mind that waste can increase the flue gases and the content of microelements in products, but also reduce the required amount of energy, those topics were secondary that came up from the main aim. Despite extensive lab-scale research on waste recycling [9-12], just a limited transfer of results to the industry occurred. This can be seen as a high-priority issue - caused by the lack of pilot plant and industrial scale trials - somehow justifying the reluctance of brick manufacturers to introduce waste for possible problems (damaging the kiln and/or overpassing emission thresholds and/or non-conformities of products).

On the other hand, the production of ceramic materials implies considerable embodied energy in the finished products, and the industry still relies largely on non-renewable energy sources. Beyond the related environmental issues, the impact of fossil fuel prices on production strategies has been lately highlighted and poses a major concern for most manufacturers worldwide. In particular, the highest cost for a brick factory is undoubtedly energy (i.e., approximately 40 % by accounting only for the firing stage). Although it depends on several factors (e.g., type of fuel, firing kiln technology, raw materials characteristics, etc.), the energy performance for sustainability and competitiveness of the sector is crucial [13].

This paper provides a brief analysis of the literature and an insight into challenges that the traditional ceramic industry (particularly clay bricks, roof tiles, rustic tiles, etc.) has to face in the future and perspectives to globally enhance the sustainability of this sector, especially through circular economy.

2. State of the art and perspective of circular economy at laboratory-scale

Natural raw and waste materials combined to produce ceramics is a well-known concept, extensively researched. Materials of both organic and inorganic origin (or their mixtures) have been utilized [12]. Using waste can result in ceramics with reduced density and thermal conductivity, among other advantages. There are possible savings in energy, but also an increased carbon footprint and health risks due to the emission of volatile organic compounds. The value from the perspective of waste disposal and the decreased consumption of primary raw materials is not insignificant [14]. Numerous studies were conducted in the past to ascertain in which way the inclusion of industrial waste modifies the properties of fired bricks and roof tiles (such as microstructure, thermal conductivity, the coefficient of thermal diffusivity, water absorption, shrinkage, compressive strength, bulk density, etc.) [12, 15-18]. Examples of waste tested in fired brick include rice husks, sunflower hulls and sawdust [14], and different kinds of coal ashes [19]. Several publications related to the waste materials implementation in roof tiles have been published, for instance about aluminum sludge and glycerine pitch [20]. For a wider treatise on waste recycling in clay bricks refer to the literature [10–12,19,21,22].

When choosing a waste material that can be used to produce ceramic products, it is important to pay attention to how that addition changes the content of amorphous and crystalline matter in the final product. The increased quantity of crystalline phase is important for more porous products with water absorption above 6 % [23].

As far as laboratory examinations are concerned, more tests are needed regarding in the future regarding durability, frost resistance, leaching of potentially toxic elements, and generally aggressive environmental exposure. When designing experiments, it is necessary to keep in mind that the processing must be as close as possible to real industrial conditions [23]. Besides, the awareness of the market conditions and the requested quality of the final industrial products must not be neglected [24].

Nowadays, bricks are increasingly used as a basis for attaching ventilated façades. Therefore, thermal and acoustic insulation behaviors are required. In this respect, the use of certain waste materials can reduce thermal conductivity by increasing the porosity of fired products. Conversely, higher porosity frequently hinders mechanical resistance, whose minimum value is limited by building codes and standards (Fig. 1).

3. State of the art and perspective of circular economy at industrial scale

The economic importance and size of the construction sector are renowned, as the efforts spent to improve its environmental impact [2, 60,61]. However, due to the intensive energy requirements for brick and tile production [1,60,61], its release of flue gases [1,2,62] and intensive raw materials consumption [63], with related quarry management and reclamation issues [63,64], this sector is called upon to further improve its sustainability.

Nevertheless, the overall environmental impact of brick and tile production needs to be assessed more carefully, bearing in mind the long service life of ceramic products (the durability of a brick house is estimated at ~ 150 years) so that the embodied energy per year of use is much lower than with alternative building materials.

In general, for the production of ceramic tiles, recycling is a consolidated practice (Water and Solid waste management) and only a small quantity of hazardous waste (spent lime) is sent to landfill [61].

From the literature, it appears that only a few types of waste and by-products have the potential to be successfully introduced into industrial practice. For instance, 10 % of fired brick scrap can be used as a substitute for sand in illite-chlorite carbonate-rich clay (Fig. 2), but with no frost resistance [65]. Besides, packaging and cathode ray tube waste glasses were tested and gave promising results [66]. The only additive of organic nature implemented in industrial-sized bricks was coal dust [8], which is added to an illitic loess clay in an optimal amount of 3 %.

The brick industry has used different coal ashes for a long, but the quantities are not often disclosed. Among the published studies, most are oriented toward producing industrial-sized hand-molded samples, where determined optimal quantities are up to 50 % [16]. Other successful examples are the incorporation of rice husk and sugarcane bagasse ashes into a soil containing low clay minerals quantity [67], and rice husk ash into a high clay mineral raw material with a significant quantity of iron [68]. In the roof tiles industry (Fig. 2), examples of waste recycling at industrial scale include 20 % of ceramic sludge [69], <15 % of rice husk ash [70] (and a combination of ceramic sludge and rice husk ash [71], and sewage sludge [72].

Although experimental studies on the implementation of secondary raw materials have been done extensively by universities and research institutes in the past, technology transfer and results on industrial-sized products are scarce and more research and development is needed. In this sense, the brick industry is facing new challenges, such as the ecodesign of new construction products in line with the new trends in construction (e.g., larger low-density prefabricated products for industrialized settings, which facilitates the dismantling of buildings and reduces the amount of waste generated). Improvements in the formulation of the raw materials are also required for more effective drying and firing stages, which necessitates extensive study [1,66]. In this framework, the contribution from computational tools that have been developed by laboratory experiments is fundamental to improving the selection of waste materials and strengthening the knowledge of the influence of the various parameters involved [12,24].

Types of waste long used in industrial practice include [73]:

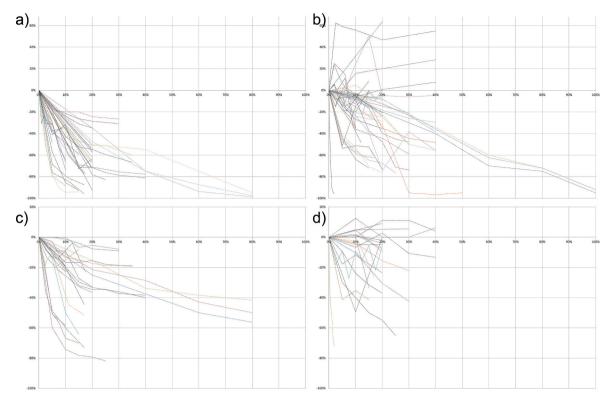


Fig. 1. Relative variations for compressive strength by adding organic (a) and inorganic (b) wastes and for thermal conductivity by adding organic (c) and inorganic (d) wastes, by literature [25–59].

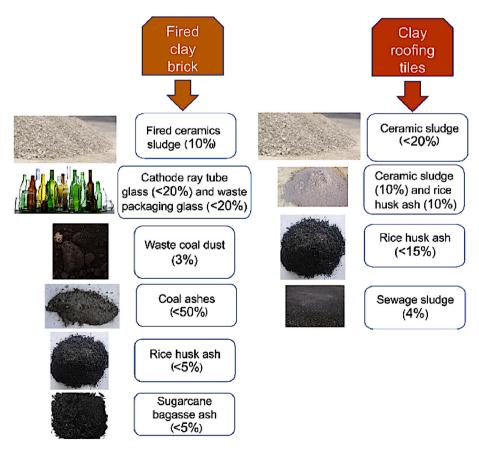


Fig. 2. The successful waste implementation in traditional ceramic masses for industrial prototypes.

agricultural residues – like olive pomace sludge and olive stone (0.2–6.0 wt %), biomass ash (1.0 wt%), grape seeds (0.8 wt%), rice husk – and industrial residues, such as coal fly ash, sawdust, paper sludge, petroleum coke, soil from construction earthworks, and others (generally 2–9 wt %). Heavy hollow blocks (so-called 'thermal blocks', where porosity is a value to improve thermal resistance) are usually the main goal of waste recycling. To a lesser extent, also common bricks and hollow bricks and blocks can contain recycled materials. No waste is commonly incorporated into higher value-added products (e.g., roof tiles, large hollow slabs, facing and paving bricks and tiles).

4. Challenges to improve sustainability of the ceramic industry

Together with raw materials, energy demand and gaseous emissions are critical factors for the ceramic industry. According to life cycle analysis, the firing stage in this industry has the greatest environmental impact [61,74]. Apart from the best available techniques and proper process management [1], the main issue is globally the type of fuel used, which is then related to the composition and quantity of flue gases. As the most extreme example, petroleum coke is still used in developing countries, which increases $\rm CO_2$ emissions from the furnace, causes corrosion of refractory materials, pipelines and metal structures, along with sulfate efflorescence. Environmental pollution will undoubtedly be reduced as a result of improved energy efficiency, the development of cleaner technologies, and the utilization of renewable energy sources [1, 2,75,76].

In the traditional ceramic industry, 50 %–88 % of the total $\rm CO_2$ emission comes from fuel, depending on the type of product [1]. Specifically, annually, in the EU the production of wall and floor tiles, bricks and roof tiles, and refractories results in 19 Mt of $\rm CO_2$ emissions [How to decarbonize the ceramics industry [77].

For example, the highest emission was recorded in the case of roof tiles, and the lowest in porous blocks [77]. To reduce CO2 emissions, emphasis is placed on the use of alternative fuels, even though most cannot be considered carbon neutral (i.e., the amount of ${\rm CO_2}$ released by combustion and the amount absorbed by photosynthesis are in balance). These include biomass from agriculture and forestry, and biodegradable urban, animal and paper waste. However, additional research is required on this topic, since biomass combustion can involve health risks. Today, the ceramic industry in Europe uses natural gas (85 % of the total energy demand) and electricity (for the remaining 15 %) in the production process [1]. In many developing countries, coal is still the predominant choice. Switching to the use of electricity for firing would move upstream the generation of CO2 (thermal power plants) and increase the production costs [1]. Efforts to "decarbonize" energy will reduce indirect electricity emissions from the ceramic industry but are estimated to be insufficient for the 2050 target. Future alternatives include the use of green hydrogen for the drying and calcination stages, although its adaptation to the ceramic industry is expected to be in the medium to long term, as well as the implementation of CO2 capture technology. Still in the shorter term, biofuels should be implemented as a source of thermal energy [78]. The cost of adaptation to the new energy supply will have a big impact on how competitive the ceramic industry will be globally. The additional use of solar energy would currently be the most beneficial option in terms of carbon footprint. Using locally accessible waste materials without carbonates can greatly reduce clay-related CO2 emissions, which are now estimated to 17 % [2]. Nonetheless, carbonates play a beneficial role during firing, increasing the mechanical performance of porous products, and investigation on the cost-benefit of their replacement is needed.

To establish a comprehensive strategy based on resource efficiency, it must be taken into account that future trends imply the need to reduce ${\rm CO}_2$ emissions in a sector with high-energy demand for drying and firing, maintaining brick quality and applying circular economy and eco-design criteria in processes and products.

5. Conclusions

The strategic goals of the sustainable development of ceramic materials production will shift to a competitive, decarbonized and resource-efficient economy, and the maintenance of a high and stable economic standard and employment growth. The ceramic sector is important for Europe's 2050 decarbonization target, and it must expand current knowledge and expertise to meet this great challenge and develop novel ground-breaking technologies. To put the principles of the circular economy into practice, future trends in this sector must address the following:

- Improving laboratory research, bringing experimental conditions as close as possible to industrial conditions,
- 2. Utilizing renewable energy sources as fuel (green hydrogen, biofuels and decarbonized electricity),
- 3. Developing CO₂ capturing technologies,
- 4. Open reporting on results, with mandatory highlighting of negative conclusions and problems/risks and opportunities,
- Enhancing industry-academia communication, promoting the publication of results,
- Intensifying usage of mathematical tools and computational approaches.

Author's contributions

All authors contributed equally to the work.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Availability of data

Data sharing does not apply to this article as no new data were created or analyzed in this study.

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