



# Use of industrial ceramic sludge in brick production: Effect on aesthetic quality and physical properties



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## HIGHLIGHTS

- Re-using sludge from the ceramic industry as an alternative eco-friendly additive.
- Aesthetic quality and physical properties of traditional bricks and new mix design.
- Economic and ecologic ways of developing bricks from recycled waste.

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## ABSTRACT

Most brick companies nowadays focus their research on the recycling of waste, in order to be able to market new types of bricks. In this work, we explored the possibility of using ceramic sludge in brick production, aiming to find an alternative eco-friendly additive to produce “eco-bricks” characterised by suitable mechanical and aesthetic properties and durability. For this purpose, two types of bricks produced by an Italian factory (SanMarco-Terreal) were compared with a newly designed brick obtained from the same starting clay, with the addition of ceramic sludge in place of the traditionally used siliceous sand. Bricks and raw materials were investigated with a multivariate approach, consisting in the mineralogical and chemical analysis, and the final products microstructurally investigated and their physical-mechanical properties determined. Results show that bricks produced with added ceramic sludge can substitute traditional bricks well, fulfilling aesthetic requirements and maintaining sufficient mechanical properties. However, one drawback was that these new materials did not respond to freeze-thaw cycles, highlighting their potential vulnerability in cold climates.

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## 1. Introduction

The raw material used in brick production is mostly composed of clay. The growing demand for high-quality final products, the need to expand company production, and increasing attention to environmental problems (cfr. many regional, national and EC norms) have led to the use of several additives in addition to normal raw clay materials. Additives, both natural and synthetic, act as auxiliary “raw materials” and influence many properties of fired products, such as colour, mechanical strength and durability [1]. In ordinary brick production, quartz-rich sand is the additive mainly used as temper (10–20 wt%) since it is easily available, does not release pollutants at any stage of the production process, reduces

plasticity, prevents shrinkage, and improves the mechanical resistance of the finished product.

Several researches have been carried out in the last few decades on creating new types of bricks using additives, often consisting of residual urban and industrial materials [2–9]. These works have often demonstrated the potential of these materials, which have technical advantages and can reduce the environmental impact. The introduction of industrial waste in brick production is also a response to the problem of the disposal of large amounts of waste materials from various industrial activities. Storing these waste materials and the resulting global environmental hazard has increased the demand and development of sustainable alternatives. For these reasons, industrial and academic attention has focused on developing environmentally friendly, low-cost and lightweight construction materials obtained from waste. Bricks mainly consist of clayey materials, can tolerate the addition of waste even in significant percentages, and are therefore suitable for this type of re-use [1].

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One type of waste is sludge from ceramic production, consisting mainly of silico-aluminous-based components (>50%), generally with low contents of heavy metals [1]. Being compositionally similar to the raw clayey materials of bricks, this type of waste is related to ceramic production and often inadequately disposed of, although it is an important source of additives in brick production if properly designed new mixes are created. Nowadays, most ceramic-producing companies have addressed their research and organisation of productive plants to recycle waste, save energy and re-use resources (mainly water) deriving from the production cycle, thus saving natural resources and resolving the waste disposal problem. All these aspects contribute much to the evaluation of company performance. Although re-using ceramic production waste (shards) is quite common, re-using sludge is more difficult, due to its intrinsic nature. Sludge is very fine-grained and, according to the type of ceramic production from which it derives, does contain heavy metals in variable quantities, from few ppm as they naturally occur in the base clay and possibly added temper, to some percent, as in the case of glazed and colour ware. Therefore, a sludge does not reflect environmental and human safety requirements, indicating that continual quality control of used materials is always important.

This work explores the possibility of using ceramic sludge in brick production. Two types of commercial bricks produced by the Italian factory SanMarco-Terreal were analysed, and the results were compared with a newly designed brick obtained from the same starting clay tempered with ceramic sludge instead of the traditional siliceous sand. More in detail, the sludge here used was obtained from the mechanical treatment (such as lapping or scrapping) of a clay based highly-fired ceramic material. The mineralogical composition, texture, physical properties, water behaviour and durability of both traditionally produced and newly designed bricks were analysed by a multi-analytical approach, and the possibility of recycling ceramic sludge in brick production was critically evaluated.

This research arose from definite interest on the part of the company, which provided full technical support.

## 2. Materials and methods

Three types of bricks (two (bricks 1 and 2) commercialized and industrially produced, and one (brick 3) experimentally produced in the SanMarco-Terreal laboratory, simulating the same preparation, forming and firing conditions used for the industrial ones) were prepared with the same carbonate-rich clay, quarried from the area of Marcon (Venice inland) and fired at 1050 °C. Any differences among them were due to the additives used. Bricks were formed by the soft-mud shaping method, in which the clay paste is placed into 5 × 12 × 20 cm moulds, the walls of which are spread with quartz sand to avoid clay from sticking and favour both water drainage while pressing and the separation from the mould. The colour of the commercial bricks was yellow (brick 1) and black (brick 2), and both were prepared by tempering clay with 10 wt% of siliceous sand, but the black brick was obtained adding to the clay paste and quartz sand also 15 wt% of the colouring agent hausmannite (Mn<sup>2+</sup>Mn<sub>3</sub><sup>3+</sup>O<sub>4</sub>). Hausmannite is a secondary waste product, approved according to valid environmental impact criteria [10] and derived from industrial sources, such as the production of ferroalloys and MnO<sub>2</sub>-based batteries. The newly designed brick (brick 3) was prepared by tempering the same clay with 10 wt% of dried ceramic sludge provided by SanMarco-Terreal.

Raw materials (clay and sludge) and fired products were chemically analysed by X-ray Fluorescence (XRF) using a S4 Pioneer (Bruker AXS) spectrometer (estimated detection limit for major elements: 0.01 wt%); ZAF correction was performed systematically

[11], and the NCSDC 74301 (GSMS-1) standard [12] was applied. Grain-size distribution in the size range between 0.02 and 2000 μm of the sludge was determined using a Mastersizer 2000LF laser diffraction particle size analyser (Malvern Instruments).

The mineralogical composition of raw materials and bricks was determined by X-ray Powder Diffraction (XRPD) on a PANalytical X'Pert diffractometer with Co-Kα radiation, operating at 40 kV and 40 mA intensity; XRPD data were interpreted by X'Pert PRO HighScore Plus<sup>®</sup> software (PANalytical). Semiquantitative phase analysis was performed on the crystalline phases using the RIR method. Since none internal standard was used during the XRPD acquisition, relative abundance of the amorphous phase was expressed only qualitatively, taking into consideration the pattern profile.

Colour of the raw materials and the fired bricks was determined on a Konica Minolta CM-700d spectrophotometer (10 measures per samples were performed). According to the CIE system, colour is described considering a parameter for luminescence (L\*) and two chromatic coordinates, a\* and b\*, which reflect the amount of red-green (-60: green,+60: red) and yellow-blue (-60: blue,+60: yellow), respectively. Colorimetry was performed in dry and then in wet conditions on the fired bricks, to determine all possible colour changes due to the presence of water or humidity. Colour difference ΔE was calculated according to the following equation:

$$\Delta E = \sqrt{(L_1^* - L_2^*)^2 + (a_1^* - a_2^*)^2 + (b_1^* - b_2^*)^2}$$

where subscript 1 refers to measurements on dry samples and subscript 2 on wet ones.

Petrographic features, texture and degree of vitrification of the bricks were studied on polished thin sections by Optical Microscopy (OM) under polarized light with an Olympus DX-50 equipped with a Nikon D7000 digital microphotography system, and on back-scattered electron (BSE) images obtained by Field Emission Scanning Electron Microscopy (FESEM) with a LEO GEMINI 1530, coupled to an INCA-200 Oxford microanalysis system.

Water absorption [13] and drying [14] tests on fired bricks were performed on cube-shaped samples (50-mm edge) on three samples of each brick type. Free and forced absorption (A<sub>f</sub> and A<sub>f</sub>), drying index (D<sub>i</sub>), apparent and real density (D<sub>a</sub> and D<sub>r</sub>), open porosity (P) and degree of pore interconnection (A<sub>x</sub>) were calculated.

Capillary rise (B) was studied on three prism-shaped samples (25 × 25 × 120 mm) for each brick type, according to UNI EN 1925 [15]. The coefficient of capillarity (K<sub>s</sub>) was calculated 9 min after the test began.

The distribution of pore access size (range 0.003–360 μm) was determined by Mercury Intrusion Porosimetry (MIP) on a model 9410 Micromeritics Autopore apparatus, which can generate a pressure of 414 MPa. Freshly cut samples of approximately 2 cm<sup>3</sup> were oven-dried for 24 h at 110 °C and then analysed. Nitrogen adsorption was used to determine porosity in the range diameter 2–3000 Å. Sorption isotherms were determined at 77 K on a Micromeritics Tristar 3000 in continuous adsorption conditions. Prior to measurement, samples were heated to 130 °C for 24 h and out-gassed to 10<sup>-3</sup> Torr on a Micromeritics Flowprep. The Barrett-Joyner-Halenda (BJH) method was used to obtain pore-size distribution curves.

V<sub>p</sub> (compression pulse) and V<sub>s</sub> (shear pulse) propagation velocities were measured to check the elastic-mechanical characteristics and structural anisotropy of the fired bricks, and to detect compactness variations during and after ageing. Waves were transmitted in the three perpendicular directions of the cube-shaped samples (50-mm edge) on a Panametrics HV Pulser/Receiver 5058PR coupled to a Tektronix TDS 3012B oscilloscope.

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