Valorisation of GRP Dust Waste in Fired Clay Bricks

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In Europe, the total amount of Glass Reinforced Plastic (GRP) waste is increasing. In order to valorise GRP dust (GRPd) waste and to reduce the consumption of nonrenewable resources in building materials, GRPd has been already investigated in cementitious materials where it gives even an improvement in some performances of the final products. Valorisation of GRPd waste in the production of bricks can be considered as a further alternative. In this paper, GRPd waste was substituted to the clay volume at 5% and 10% for the manufacturing of fired clay bricks. All specimens were subjected to a firing temperature of 850°C for 6 hours, then tested and compared in terms of porosity, compressive and flexural strengths, density, and water absorption. Despite a decrease in compressive strength up to 46% with 10% of GRPd substitution and an increase of water absorption from 14% to 29% with 5% and 10% of GRPd substitution, respectively, an increase in terms of lightness (about 10%), maximum flexural strength (up to 31%), and deflections at the maximum load (up to 130%) has been registered by specimens with 10% of GRPd substitution.

1. Introduction

The building construction sector produces more than 40% of the world’s energy consumption and one-third part of the related greenhouse gas (GHG) emissions [1, 2]. In 2014, only in Europe, constructions and construction works have contributed to a global CO₂ emission of 8.7%, equivalent to 627 kg of CO₂ per inhabitant [3]. Moreover, it is estimated [1] that the sector contributes to 30% of raw material extraction, 25% of solid waste generation, 25% of water use, and 12% of land exploitation.

Henceforth, for reducing construction sector impacts, the development of more environmentally friendly building materials must consider not only operational requirements (e.g., thermal properties, mechanical performance, and water absorption) [4, 5], but also their manufacturing process (e.g., energy consumption, GHG emissions, and amount of raw materials).

Fired clay bricks (FCBs) are one of the most produced construction materials [6]. Every year, approximately 1500 billion bricks are manufactured worldwide, involving more than 4000 million tons of clay extracted to be processed by more than 100,000 kilns [7]. The extensive use of natural clay deposits for bricks production has bring into focus an alarming deficit level of this material [8] so much that some countries, like China, have limited the use of bricks made from clay [9] and force researchers to study new ways for their production.

In the construction materials related research, the most common practice consists in replacing natural nonrenewable raw materials with wastes and industrial by-products [10–19]. In this way, not only an added value to the waste, contributing to the circular economy, is ensured, but also some performances of the final material can be improved.

In this line, several studies are already reported on the manufacturing of FCBs by including wastes/by-products, such as biomasses or biomass ashes [20, 21], glass wastes [22, 23], and industrial by-products [24, 25]. Results have shown that the addition of organic residues always reduces compressive strength, bulk density, and thermal conductivity but increases the water absorption of bricks. On the contrary,
inorganic residues affect these properties depending on their compositions, since metals and minerals introduced by these additions react and melt with the clay minerals in very different ways [6, 26].

Besides the clay issue, in Europe, approximately 1069 million tonnes of Glass Reinforced Plastic (GRP) have been manufactured in 2015 and the production will increase in the following years, according to the European Composites Industry Association (EuCIA) [27]. In 2015, the total amount of end-of-life and production wastes generated by glass thermoset composites market in Europe has been estimated to reach 304,000 tonnes [28], triggering the interest in optimizing the GRP waste recovery.

Nowadays, landfill as nonhazardous waste remains the most popular solution to manage GRP waste for the difficulty of separating the different parts, its intrinsic thermoset composite nature, and the insufficient knowledge on recycling options.

The partial replacement of aggregates with GRP dust (GRPd) waste in concretes and mortars at high dosages worsens the mechanical performances [29–31]. However, at low dosages (up to 10% of sand volume), it increases workability, flexural strength, deformability, and thermal insulation properties, and it decreases capillary water absorption, efflorescence phenomena, and the risk of cracking due to lowered restrained shrinkage [32–34].

Similarly, the effect of GRPd waste addition on the production of bricks can also be considered. The dual nature of GRPd waste (approximately 80% polymeric and 20% glassy) could be interesting for a possible synergistic combination of properties deriving by both organic and inorganic matters: during the firing process, the polymeric part will burn increasing the matrix porosity (improvement of lightness) [35], whereas the glass fibres will partially melt and could act as reinforcing agent, decreasing in this way the fragility and improving both the bending strength and the ductility of the final product [23].

Normally, the thermal treatment is divided into three phases: heating, firing, and cooling [6]. In the literature, several firing processes have been proposed: Karaman et al. [36] fired bricks from 750°C to 1050°C for 8 hours; Mohammed et al. [37] compared performances of bricks containing organic and inorganic waste fired at three different temperatures (800, 900, and 1000°C) for about 4 hours; Dondi et al. [23] investigated bricks manufactured with glassy wastes at firing temperatures of 900, 950, and 1000°C always for 4 hours.

To the best of authors’ knowledge, the replacement of clay with GRPd waste in FCBs production is still an unexplored issue. For this reason, in the present paper, FCBs were manufactured by replacing the 0–5–10% of clay volume with GRPd. The effect of these substitutions on FCBs performances was studied in terms of porosity, compressive and bending strengths, bulk density, and water absorption.

2. Materials and Methods

2.1. Materials Characterization. FCBs were manufactured with clay supplied by a local Italian company. Firstly, the clay was dried at 105°C. Then, it was crushed and milled in order to obtain a powder with a particle size suitable to pass through a 150 μm sieve. The bulk density of the clay is 1.8 g/cm³. The mineralogical composition of clay was studied by X-ray diffraction (XRD) by means of an RX Philips PW 1730 on previously pulverised samples passing through a 75 μm sieve. As it can be observed in Figure 1, the main mineralogical phase present in the clay sample is quartz. The presence of dolomite, micas (muscovite) and, overall calcite indicates that the clay is rich in calcium. Additionally, a phyllosilicate mineral (chlorite) and a feldspar (albite) are detected in the clay sample.

GRPd, provided by an Italian shipyard, is a material classified as a nonhazardous waste, and it has an average diameter of 100 μm and a bulk density of 1.3 g/cm³. GRPd was added to bricks in two manners: (i) as it is and (ii) with a previous sieving treatment (sieve opening 250 μm) in order to increase the glassy part (Figure 2).
The SEM analysis was performed with a ZEISS 1530 SEM, equipped with an EDAX probe, operating at 10 keV. The image obtained by SEM, given in Figure 3(a), shows that the GRPd morphology is characterised by polymeric granules surrounding low alkali glass with 0.02–20 mm lengths. EDAX composition shows the presence of calcium and a high amount of silicon, mainly due to the glassy part of the sample (Figure 3(b)) [33].

DT/TG analysis with a METTLER TOLEDO TGA/SDTA 851, equipped with a TSO 800GC1 programmable gas switch from 30 to 1000 °C, with 10 °C/min heating rate of GRPd before and after sieving is performed in air (Figure 3(c)). The polymeric part degrades mainly in two steps at $T = 340^\circ C$ and $T = 490^\circ C$. At $T > 550^\circ C$, all the polymeric part of GRPd is burnt, and the residual mass is formed only by the glassy part. DT/TG results show that GRPd is composed by 20% of glass fibres and 80% of polyester resin by volume. In the case of the sieved GRPd sample, the volumetric percentages change to 30% and 70% for glass fibres and polyester resin, respectively.

2.2. Preparation of Bricks. Bricks were obtained by moulding process. In order to reach a good plasticity, a predetermined weight of the dry clay was mixed with 23% of water by clay mass. Specimens without GRPd addition, labelled as STD, are the reference. GRPd was substituted to the clay at two different volumetric dosages: 5% and 10% (related specimens are labelled VR 5 and VR 10, respectively) keeping constant the water amount. At the dosage rate of 5%, GRPd was also added with a previous sieving treatment (sieve opening 250 μm), and related specimens are labelled as VR 5 siev.

The obtained body was pressed (with a pressure of 1 MPa) into a cubic or prismatic mould of $5 \times 5 \times 5$ cm$^3$ and $4 \times 4 \times 16$ cm$^3$. For each mixture, three specimens per test were prepared. The specimens were initially dried at room...
temperature (20°C) for 24 h and then oven heated at 105°C for 24 h in order to prevent specimen bloating or swelling related to water evaporation that could cause expansive phenomena during the firing process [38].

Firing was done in an electrically operated muffle furnace. In the current study, bricks were fired up to 850°C, for 8 hours: 2 hours are implied for heating and 6 hours for firing. After firing, the muffle furnace was switched off, and the specimens were allowed to cool down until room condition (20°C). The firing temperature of 850°C was chosen since this relatively low temperature permits a lower energy demand for the manufacturing process by ensuring nonetheless acceptable mechanical properties of bricks.

2.3. Characterization of Bricks

2.3.1. Chemical Characterization. The mineralogical characterization of brick specimens was carried out by XRD analysis on samples sieved at 75 µm obtained by pulverising bricks after the thermal treatment.
2.3.2. Physical Characterization. The bulk density was calculated by measuring the weight and the volumetric dimensions of specimens previously dried at \( T = 105^\circ C \) until constant weight was reached.

Fired clay bricks are porous media; hence, the pore structure was investigated by mercury intrusion porosimetry (MIP) analysis with a Thermo Fisher 240 Pascal, operating between 0.1 MPa and 200 MPa pressures. For each composition, three small fragments were tested, and the average results are reported.

Water is the medium and the main carrier of aggressive ions that can cause degradation of construction materials [39, 40]. Furthermore, porous media are more susceptible to salt crystallization and freeze-thaw cycles. For this reason, the study of water absorption is considered to be of primary importance to stand the durability of a building material. In this experimentation, water absorption was evaluated on three cubic specimens (5 \( \times \) 5 \( \times \) 5 cm \(^3\)) for each FCB typology, previously dried at 105 \(^\circ\) C in an oven. The specimens were weighted before and after 24 h of total immersion in deionized water. Average results are expressed in terms of percentage of water absorbed according to EN 772-21 [41] but with different size of specimens.

2.3.3. Mechanical Characterization. Mechanical properties of specimens (compressive and bending strengths) were investigated by using a “Galdabini” hydraulic press with a precision of 1\% at a loading speed of 0.3 (N:mm\(^2\))/s according to EN 772-1 [42], but with different sizes of specimens. Compressive strength was studied on three cubic specimens of 5 \( \times \) 5 \( \times \) 5 cm \(^3\). Three point-bending strength and the relative vertical deflection at the maximum flexural load were measured on prismatic specimens (4 \( \times \) 4 \( \times \) 16 cm \(^3\)).

3. Results and Discussion

3.1. Chemical Characterization. Firstly, the chemical analysis of fired clay bricks with and without GRPd has been investigated. It is well known that several properties of FCBs are highly influenced by the clay composition [43]. Indeed, not only the temperature, but also the bulk composition of the original clay (Figure 1) are the driven forces in the formation of different phases.

As Figure 4 clearly shows, gehlenite is detected after firing the Ca-rich clay. Gehlenite usually starts to form at 850 \(^\circ\) C, such as in this case, from the decomposition of clay minerals and calcite, which decomposes at 650 \(^\circ\) C. The transformation of dolomite usually takes place at 800 \(^\circ\) C, so that in both bricks it is no more detected; on the contrary, albite and muscovite are still visible because they do not incur into modifications at the low firing temperature of the present work [44, 45].

The addition of GRPd does not modify the mineralogical composition of FBCs. Even if VR 10 is manufactured with the highest amount of GRPd, and thus with highest percentage of glassy phase, no marked raise in the background of XRD, identifying the presence of an amorphous compound, is detected. This is due to the low content of glass (2\% on the total volume of brick) in the specimens.

3.2. Physical Characterization. The firing of Ca-rich clay usually produces highly porous ceramic materials owed to the decomposition and release of CO\(_2\) during the firing process [46]. The porosimetric characteristics of FCBs are shown in Figure 5 and Table 1.

The addition of GRPd implies only a very slightly increase in total open porosity, whether if sieved or not. However, GRPd addition mainly affects the pore size distribution, shifting the curves to larger pore sizes. In this experimentation, median and modal pore diameters move from 0.492 and 0.572 \( \mu \)m to 0.581 \( \mu \)m and 0.734 \( \mu \)m, respectively, for samples without GRPd (STD) and after the substitution of clay with 5\% of GRPd (VR 5), respectively. The formation of pores with larger radii is caused by the
decomposition of the GRPd polymeric part, which consists in the major component of the waste, during the firing process. The glassy part of GRPd, even if sieved, is not sufficient to modify substantially the total open porosity values.

The microstructure affects the physical properties of specimens, such as water absorption (WA). By increasing the content of GRPd waste, also the WA increases from 21% in specimens with 0% of GRPd (STD) to 27% in those containing 10% of GRPd (VR 10) (Figure 6(a)); in fact, the higher the pore dimensions, the higher the WA value. Moreover, also the organic part of GRPd waste influences the WA: the higher its content in percentage, the higher the water absorption of bricks [26]. During the firing process, by burning, the polymeric part of GRPd produces higher open porosities, so nearly the pores are able to be connected, implying the water to be transported from the external surface to the internal locations by capillary suction [6].

Obviously, the pore size distribution and total porosity affect also the bulk density of specimens (Figure 6(b)) that shows the same trend of water absorption. The lower density value of GRPd compared to that of clay and the additional decomposition of polymeric part of GRPd during the firing process implies a decrease in the density of the final product [37] from 1500 to 1400 kg/m³ because of the volumetric substitution of the natural raw material with the waste.

3.3. Mechanical Characterization. Results obtained by mechanical characterization are shown in Figure 7.

At the same firing temperature, the mechanical properties are strongly related to the crystalline phases’ content and directly related to the percentage of calcium carbonate and calcium silicoaluminates in the matrix [46]: despite the low temperature of firing (850°C), the presence of crystalline phases, such as gehlenite, provides high mechanical strength
The addition of GRPd waste in FCBs production decreases the compressive strength from 26 to 18 MPa (30% reduction) and 14 MPa (46% reduction) with 5% and 10% of GRPd, respectively, whether if previously sieved or not.

However, bending strength increases from 5.1 to 5.9 and 6.7 MPa with 5% and 10% GRPd, respectively, whereas the corresponding deformations at the maximum load (deflection) increase by 30% and 130%, respectively. When a 5% of sieved GRPd is added, flexural strength is 6.2 MPa with a deformation at maximum load of 90% higher than that of specimens with 0% GRPd addition.

Calculating the volume of glass on the total volume of bricks, in VR 5, VR 5 siev, and VR 10 specimens, the respective volumetric glassy fractions amount to 1%, 1.5%, and 2%. Dondi et al. [23] reported a beneficial effect by using waste glass in FCBs at 2% addition by weight of clay, whereas at higher rates (5% by weight), a loss in performance was detected. In this work, a very good linear correlation between the flexural strength of bricks and their glassy content as percentage by volume, due to GRPd addition, is found (Figure 8). The use of GRPd waste addition at 10%, entailing an inclusion of 2% of glass in volume, is enough to increase the bending strength by 31% compared to the reference. Probably, glass fibres during the firing at 850°C have partially melted playing the role of sintering promoters through the formation of a viscous phase and ensuring a higher bending strength and a higher deflection during the operating conditions [23, 47]. The higher bending strength and deflection of GRPd bricks, despite the lower compressive strength, can guarantee the production of bricks with lower crack tendency not only during their service life but also during handling and transportation processes which generally break a high number of bricks [38].

4. Conclusions

The inclusion of glass reinforced plastic dust (GRPd) waste as partial replacement of the natural clay for the production of more environmentally friendly fired clay bricks (FCBs) has been investigated. GRPd was substituted at 5% and 10% by volume of clay, as it is and after a previous sieving in order to enrich the glassy fibres content.

Results demonstrate that GRPd addition in FCBs production not only valorises GRPd waste and increases sustainability of FCBs, but can also lead to an improvement of some final performances of bricks.

In particular, the following have been found:

(i) GRPd waste addition does not modify the mineralogical composition of FCBs at these low dosages.

(ii) GRPd waste addition slightly increases the total porosity and mainly affects the pore size distribution, shifting the curves to larger pore sizes. The sieving process does not significantly affect this property.

(iii) GRPd waste addition increases the water absorption from 21% in specimens with 0% of GRPd (STD) to 24% and 27% in those containing 5% and 10% of GRPd, respectively, but decreasing the bulk density from 1500 to 1400 kg/m$^3$, it increases the lightness of the final product.

(iv) GRPd waste addition reduces the compressive strength of FCBs of the reference from 30% to 46% reduction with 5% and 10% of GRPd, respectively, but it increases the bending strength of the reference by 16% and 22% at 5% addition rate as it is and sieved, respectively, and by 31% at 10% addition rate. The corresponding deflections are enhanced by 30%, 90%, and 130%, respectively. The higher bending strength and deflection of GRPd bricks, despite the lower compressive strength, can guarantee the production of bricks with lower crack tendency not only during their service life, but also during handling and transportation processes, which generally break a high number of bricks.

(v) A good linear correlation between the flexural strength of FCBs and their content of glass, expressed as volumetric percentage, has been found.

Conflicts of Interest

The authors declare that there are no conflicts of interest regarding the publication of this paper.

References


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