sustainability

MDPI

Article

Crushed Bricks: Demolition Waste as a Sustainable Raw Material for Geopolymers

Gigliola D'Angelo [1,2], Marina Fumo [1], Mercedes del Rio Merino [2], Ilaria Capasso [3,*], Assunta Campanile [4], Fabio Iucolano [4], Domenico Caputo [4] and Barbara Liguori [4]

1 Department of Civil, Building and Environmental Engineering (DICEA), University of Naples Federico II, Via Claudio 21, 80125 Naples, Italy; gigliola.dangelo@unina.it (G.D.); marina.fumo@unina.it (M.F.)
2 Departamento de Construcciones Arquitectónicas y su Control, Escuela Técnica Superior de Edificación, Universidad Politécnica de Madrid, Avda. Juan de Herrera 6, 28220 Madrid, Spain; mercedes.delrio@upm.es
3 Department of Engineering and Geology, University of Chieti-Pescara "G d'Annunzio", Viale Pindaro 42, 65122 Pescara, Italy
4 ACLabs-Applied Chemistry Labs, Department of Chemical, Materials and Industrial Production Engineering, University of Naples Federico II, P.le Tecchio 80, 80125 Naples, Italy; assunta.campanile@unina.it (A.C.); fabio.iucolano@unina.it (F.I.); domenico.caputo@unina.it (D.C.); barbara.liguori@unina.it (B.L.)
* Correspondence: ilaria.capasso@unich.it

Abstract: Demolition activity plays an important role in the total energy consumption of the construction industry in the European Union. The indiscriminate use of non-renewable raw materials, energy consumption, and unsustainable design has led to a redefinition of the criteria to ensure environmental protection. This article introduces an experimental plan that determines the viability of a new type of construction material, obtained from crushed brick waste, to be introduced into the construction market. The potential of crushed brick waste as a raw material in the production of building precast products, obtained by curing a geopolymeric blend at 60 °C for 3 days, has been exploited. Geopolymers represent an important alternative in reducing emissions and energy consumption, whilst, at the same time, achieving a considerable mechanical performance. The results obtained from this study show that the geopolymers produced from crushed brick were characterized by good properties in terms of open porosity, water absorption, mechanical strength, and surface resistance values when compared to building materials produced using traditional technologies.

Keywords: eco-sustainable materials; construction and demolition waste; crushed brick waste; geopolymers; waste reuse

check for updates

Citation: D'Angelo, G.; Fumo, M.; Merino, M.d.R.; Capasso, I.; Campanile, A.; Iucolano, F.; Caputo, D.; Liguori, B. Crushed Bricks: Demolition Waste as a Sustainable Raw Material for Geopolymers. *Sustainability* **2021**, *13*, 7572. https://doi.org/10.3390/su13147572

Academic Editor: Mariateresa Lettieri

Received: 13 May 2021
Accepted: 1 July 2021
Published: 6 July 2021

Publisher's Note: MDPI stays neutral with regard to jurisdictional claims in published maps and institutional affiliations.

Copyright: © 2021 by the authors. Licensee MDPI, Basel, Switzerland. This article is an open access article distributed under the terms and conditions of the Creative Commons Attribution (CC BY) license (https://creativecommons.org/licenses/by/4.0/).

1. Introduction

The construction industry occupies an important position in the production of waste materials from construction and demolition activities, with around one-third of the European Union's total waste represented by this so-called Construction and Demolition Waste (CDW). Known materials range from plastics to wood and metals, but it is the inert materials category that has the highest percentage weight, reaching 75–80% of the total CDW weight [1,2]. For this reason, the EU has considered the flow of CDW as a priority stream for action. In fact, over the last decade, intense activity in the construction field in Europe has generated around 827 million tonnes of CDW on average per year, and yet only 50% of this CDW was recycled [3,4]. Moreover, climate change and environmental issues are a wake-up call to the impoverishment of resources due, above all, to the exploitation of raw materials. In this sense, the quantity of second-hand materials coming from construction and demolition operations represents a source of great value. The possibility of including CDW in building materials is, therefore, an interesting alternative.

Recently, the recycling of CDW was extensively studied and reviewed [5–8]. The data reported indicate that CDW can be successfully used in construction material production to obtain products comparable with those produced using natural raw materials.

Approximately 45% of CDW is attributed to ceramic products such as bricks, tiles, and porcelain. It is crucial to evaluate alternative applications of this waste considering the large amounts produced per year (yearly production from the EU is estimated at 855 million tonnes) [9].

Clay brick wastes (CBW) represent a valuable secondary resource for concrete manufacturing, either as a coarse or fine aggregate in concrete production, or as a supplementary cementitious material [10,11]. Recently, an interesting alternative has been reported: the development of alternative low-carbon binders (alkali-activated materials, geopolymers, and calcium sulfoaluminate cement) [12,13]. Geopolymers are obtained through a chemical reaction, starting with an alumino-silicate powder with a highly concentrated aqueous alkali hydroxide and/or silicate solution, to the production of a synthetic amorphous-to-semicrystalline alkali alumino-silicate new phase [14,15]. Alkali-activated materials (AAM) exhibit excellent properties in terms of mechanical performance [16], thermal stability [17], and durability [18].

Furthermore, geopolymers are of great interest because of their high sustainability, and reduced energy requirement in their production. In addition, the production of geopolymers allows for an 80% reduction in emissions compared to Portland cement manufacturing [14]. It has been demonstrated that any natural or synthetic material containing proper amounts of silica and alumina can be used as a precursor for the geopolymerization process [15].

Chemically bonded ceramics (CBC), belonging to the AAM class, are a group of inorganic materials that share properties with both cements and ceramics [19]. The formation of CBCs occurs via the chemical route, much like with conventional cement, while the chemical bonds and mechanical properties resemble those of ceramics. These materials allow for an alternative avenue in ceramic processing, where high temperatures are not required, and solid structures are formed in ambient temperatures in the presence of mineral impurities.

The present research aims to determine the feasibility and potential of crushed brick wastes in the production of building precast components from alkali-activation with sodium hydroxide and silicate solutions. Firstly, a complete characterization of the crushed brick waste was carried out to verify its intrinsic reactivity as a raw material for geopolymerization. Then, a ceramic product was obtained from alkali-activation of the crushed brick waste, and finally, its physical and mechanical properties were evaluated.

2. Materials and Methods

This research was divided into two experimental phases. The first phase consisted of evaluating the effective geopolymerization of the selected mixture, based on the type of waste material chosen and its percentage, through to the production of small cylindrical specimens. The second phase consisted of a scale-up of the specimens that had effectively passed the first phase, followed by the production of samples on which physical, chemical, and mineralogical analyses and mechanical tests were carried out, in order to determine the characteristics of the new material and hypothesize possible future applications.

2.1. Preparation of the Clay Brick Waste-Based Geopolymer

CBW was used as the raw material to carry out the experimental development of the research. The CBW came from the demolition site of an old building situated near the city of Naples, Italy. The original waste material (Figure 1a) was grounded to a fineness of 0.125–0.150 mm (Figure 1b). A sodium silicate solution (SS) (Na_2O, 8.15%; SiO_2, 27.40%), provided by Prochin Italia S.r.L. (Caserta, Italy), and a 10 M sodium hydroxide solution, prepared by dissolving NaOH in pellets (NaOH 98%, J.T. Baker) in bi-distilled water, were used as the alkali activators.

(**a**) (**b**)

Figure 1. Clay brick waste before (**a**) and after grounding (**b**).

Based on the clayey nature of the waste [15], the preparation of the geopolymer was carried out following the prescription reported in our previous studies [20,21]. Accordingly, powdered materials were previously dry-mixed and homogenized, and then the activator solution was added.

The alkaline activator solution was prepared by mixing sodium silicate solution (SS) with 10M sodium hydroxide solution. The weight ratio SS/N/binder was 1:1:3, and the activator/binder ratio was 0.66. Finally, the mixture was cast into cylindrical polyethylene molds (diameter 30 mm; height 70 mm) and the samples were sealed and cured for 3 days at 60 °C in an oven. At the end of the curing, all the specimens (Geo_CBW) were removed from their molds and stored at room temperature until reaching a 28-day aging period before being run for experimental analysis. A qualitative evaluation of the degree of geopolymerization was carried out by the immersion of the samples in bi-distilled water for 24 h at room temperature. Importantly, if the geo-composites were intact after the immersion period, then the geopolymer gel formation can be considered successful (Figure 2) because water absorption can be used as an indicator of the extent of the geopolymeric reaction [22].

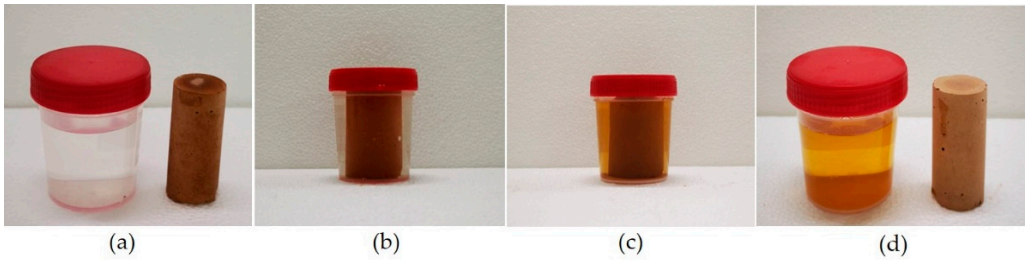

Figure 2. The geopolymeric sample (Geo_CBW) before (**a**), during (**b**), (**c**) and after immersion (**d**) in bi-distilled water for 24 h.

2.2. Mineralogical and Chemical Characterization

The chemical composition was obtained according to the following procedure. Firstly, a weighted sample was calcined at 950 °C for 2 h, then a weighted amount of the obtained sample was subjected to digestion, under microwave-induced heating (Perkin–Elmer Multiwave 3000 oven) in a standard solution obtained by mixing hydrochloric acid (37%, w/w), nitric acid (65%, w/w) and hydrofluoric acid (39.5%, wt/wt). Next, a boric acid solution was used to achieve fluoride complexation and the resulting solution was analyzed by an ICP-OES (Optima 2100 DV ICP-OES Inductively Coupled Plasma Spectrometer, Perkin–Elmer).

The mineralogical composition of the CBW was evaluated by XRD analysis of the powdered samples using a Panalytical X'Pert Pro diffractometer equipped with a PixCel 1D detector (operative conditions: CuKα1/Kα2 radiation, 40 kV, 40 mA, 2Q ranging from 5 to 80 °C, step size 0.0131° 2Φ, counting time 40 s per step).

In order to further investigate the degree of geopolymerization, FTIR and thermal analysis were carried out. FTIR tests were performed using a Nexus-Nicolet apparatus and selecting a wavenumber resolution of 4 cm^{-1} for 32 scans ranging from 4000 to 400 cm^{-1}. The FTIR spectra were collected in absorbance mode from the transparent pellet obtained by dispersing the sample powders in KBr (2% wt/wt) both with the CBW and with the geopolymeric samples produced. Thermal characterization was performed by TGA/DTGA analysis (Netzsch, STA 409 PC Luxx) in temperatures ranging from 20 to 1200 °C, with a heating rate of 10 °C/min, in a nitrogen atmosphere.

Finally, morphological analyses were carried out by SEM (SEM, Cambridge S440).

2.3. Physical Characterization of the Geopolymers

Capillary rise can be considered as one of the most significant mechanisms of water penetration into building materials, and is one of the main mechanisms responsible for building decay. For this reason, capillarity tests were performed according to the European Standard UNI EN 15801 [23], with an evaluation of the amount of water absorbed (Q) per surface unit as a function of time. Tests were performed in triplicate on cubic samples (5 cm-side). At the end of these tests, the mean value of the capillary absorption coefficient (CA, $mg/cm^2\ s^{-1/2}$) was determined. Since there is a quite linear relationship between the water adsorbed (Q) and the square root of time during shorter times, the CA value can be evaluated as the slope of the straight line in the first step (30 min) of the capillarity test [23].

The open porosity and water absorption were evaluated according to the European Standard [24]. Firstly, the specimens, after being dried at 60 ± 5 °C until a constant mass was reached, were immersed in water under vacuum at room temperature and left for two hours. Then, each sample was weighted (bulk and hydrostatic weight). The pycnometer method was used after grinding the sample to a dimension of 0.063 mm in order to evaluate the closed porosity. Each test was performed in triplicate and the results were calculated as the average values. Moreover, the water uptake from immersion, without placing samples under a vacuum, was evaluated by immersing the prismatic samples (4 × 4 × 16 cm^3) in a water tank for 48 h. Samples were first dried at 60 ± 5 °C until a constant mass was reached, then weighted before and after the immersion, and, finally, the amount of water absorbed was deduced from the weight difference.

2.4. Mechanical Characterization

The surface hardness was determined according to the standard EN 13279-2 [25]. The Shore D hardness test was carried out by establishing the indent left by an exerted force on each test specimen, measured in Shore D units varying in range from 0 (softest) to 100 (hardest).

In particular, the experimental tests were performed on the two longitudinal sides of the prismatic samples (160 × 40 mm^2) (see Figure 3).

Figure 3. Measurement of surface hardness using the Shore D test on Geo_CBW samples.

Flexural strength tests were performed in accordance with the standard EN 13279-2:2004 on the prismatic samples with the dimensions 160 mm × 40 mm × 40 mm (Figure 4a). The test machine used was the Ibertest. Test specimens were placed centrally on the sup-

porting rollers, 100 mm apart from each other. A load was applied until the specimens broke apart, while the test machine recorded the maximum load supported by the samples.

Figure 4. Performance of the flexural (**a**) and compressive strength tests (**b**) on the geopolymeric samples.

Compressive strength tests were performed following the same standard used for the flexural tests, EN 13279-2:2004 (Figure 4b). A load was then applied to the broken parts of the samples derived from the previous flexural strength tests.

The test pieces were then placed between two steel plates in a cubic geometry, with the dimensions 40 mm × 40 mm. The test specimens were loaded until rupture occurred. The compressive strength *Rc* was calculated using the formula:

$$Rc = Fc/1600 \quad (1)$$

where *Rc* is the compressive strength, *Fc* is the maximum load at fracture (N); and 1600 is the load-bearing surface (mm^2) of the tested sample. All tests were carried out in triplicate.

3. Results and Discussion

3.1. Preliminary Characterization of Clay Brick Waste

The chemical and mineralogical composition of the waste, reported in Table 1, confirmed the clayey nature of the sample with a SiO_2/Al_2O_3 ratio of 1:5 and a significant amount of alkaline and alkaline earth oxides.

Table 1. Chemical and mineralogical composition of the clay brick waste.

Major Elements (wt%)						
SiO_2	Al_2O_3	Fe_2O_3	MgO	Na_2O	K_2O	CaO
47.90	31.82	2.99	4.14	3.75	3.59	4.52
XRD mineralogical phases						
Quartz	Calcium Carbonate			Sanidine		Albite

It is worth recalling that, when clay minerals are heated at a constant rate, two principal heat effects occur: a broad endothermic effect near 550 °C, caused by the dissociation of the clay structure, and an intense exothermic peak (between 800 and 900 °C) due to the crystallization of new crystalline phases such as aluminosilicates (diopside and leucite) [26]. In addition, the XRD data from the CBW (Table 1 and Figure 5) confirmed the presence of a major crystalline phase of sodium and potassium aluminium silicates. Quartz mineral (SiO_2) and calcium carbonate ($CaCO_3$) were also detected. The amorphous aluminosilicate materials formed upon the decomposition and destuction of the clay minerals during the firing could not be observed by the XRD technique due to their amorphous nature.

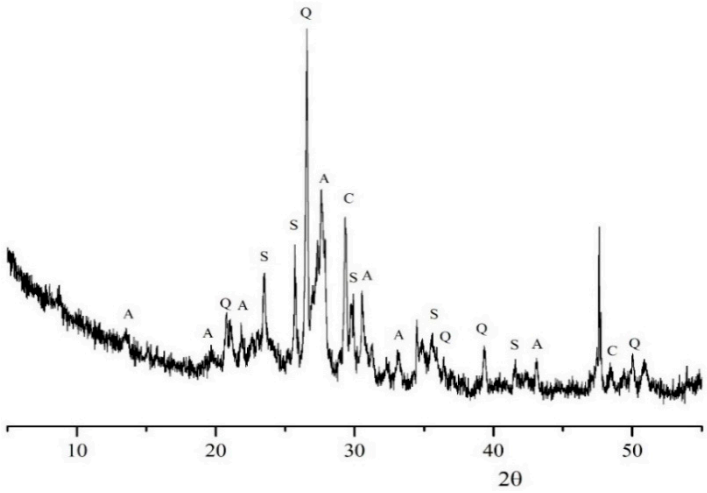

Figure 5. XRD spectra of CBW. Q = Quartz, C = Calcite, S = Sanidine, A = Albite.

Moreover, from the TGA curve of the CBW (Figure 6), a weight loss at temperatures between 100 °C and 200 °C was reported, due to the evaporation of free water absorbed by the specimen. Additionally, a small amount of weight loss (≈2%) occurred at temperatures between 650 °C and 750 °C which was attributed to the decomposition of the carbonates either present in the raw material or due to the atmospheric carbonation that occurred during the preparation of the samples prior to analysis.

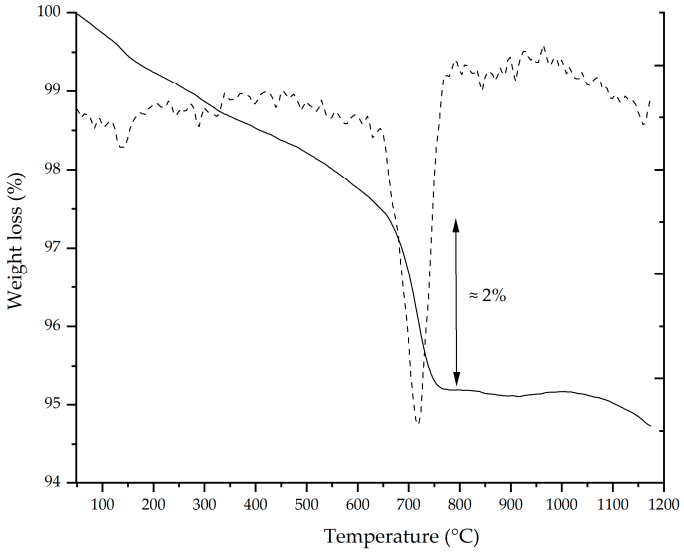

Figure 6. TGA curve (continuous line) and DTG curve (dashed line) of the CBW.

3.2. Characterization of Geopolymeric Samples

FTIR was used to verify the degree of geopolymerization of the geocomposites [27–29]. Geopolymers produced from CBW were mainly amorphous aluminosilicates themselves, and displayed FTIR spectra (Figure 7, continuous line) characterized by the typical absorption bands of Si-O-Si and Si-O-Al bonds (absorption range 600–800 cm^{-1}). The bands at 3450 cm^{-1} and 1647 cm^{-1}, associated with the O-H stretching and bending, are connected to the bound water molecules, which are surface-absorbed or entrapped in the large cavities of the molecular structure [28,30]. The intensity of these bands was greater in the FTIR spectra of geopolymers, indicating both a higher degree of water molecule adsorption in their mass and the occurrence of a geopolymerization reaction of the raw materials into a geopolymer paste [31]. Further confirmation of the presence of the aluminosilicate species typical of geopolymeric composites in Geo_CBW samples can be found

in the TGA curve and the XRD spectrum (Figure 8). In particular, the XRD spectrum (Figure 8b) indicated an appearance of peaks associated with the presence of Phillipsite ($(Ca,Na_2,K_2)_3Al_6Si_{10}O_{32}\cdot 12H_2O$), which was the only aluminosilicate crystalline phase, while the presence of other amorphous geopolymeric phases was related to the gradual weight loss detected between 250 and 600 °C in the TGA curve.

SEM investigations at different magnifications (from 50× to 3000×) of the geopolymers are presented in Figure 9. EDX spectra confirmed the silico-aluminatic nature of the geopolymeric product. The Geo_CBW sample was characterized by a porous and heterogeneous matrix with some unreacted particles (500× magnification). At higher magnifications (from 1500× to 3000×) the amorphous structure of the geopolymeric structure was evidenced.

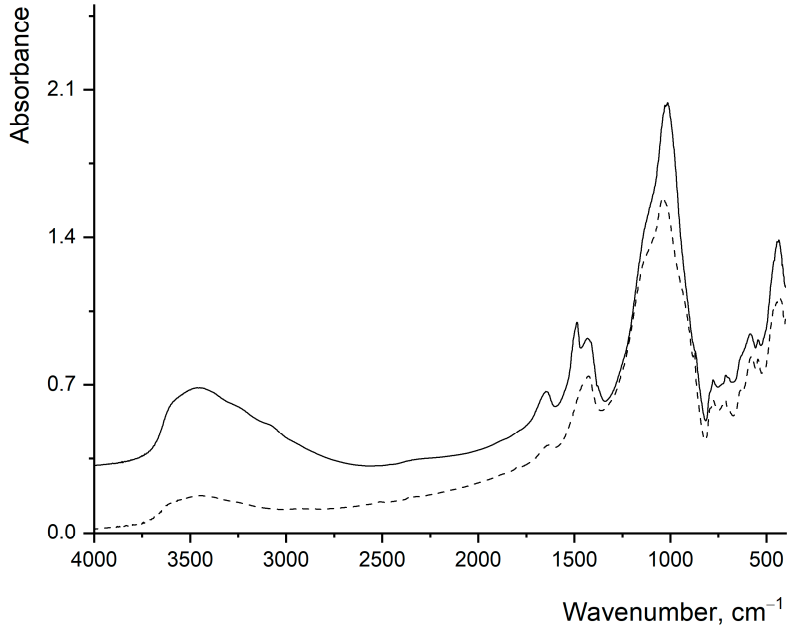

Figure 7. FTIR spectra of CBW (dashed line) and geopolymer (continuous line).

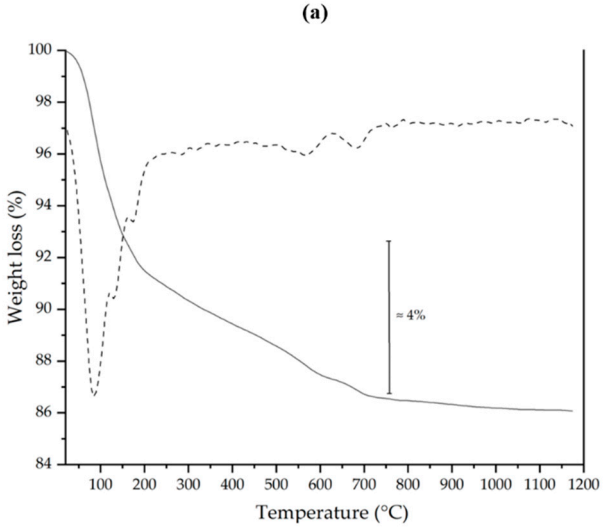

Figure 8. *Cont.*

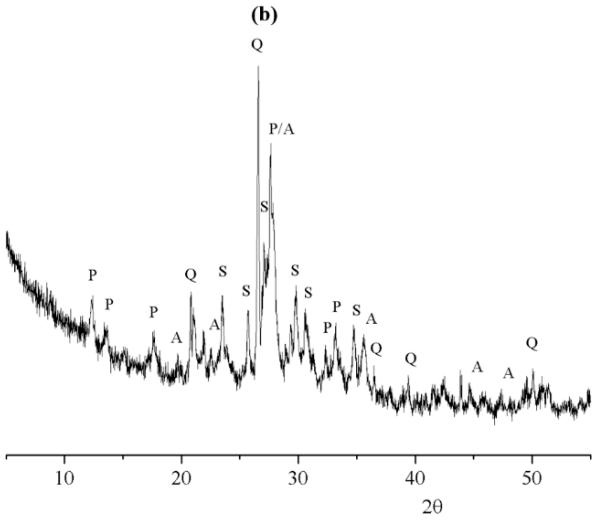

Figure 8. TGA (continuous line) and DTG curves (dashed line) (**a**) and XRD spectrum (**b**) of Geo_CBW sample. Q = Quartz, P = Phillipsite, S = Sanidine, A = Albite.

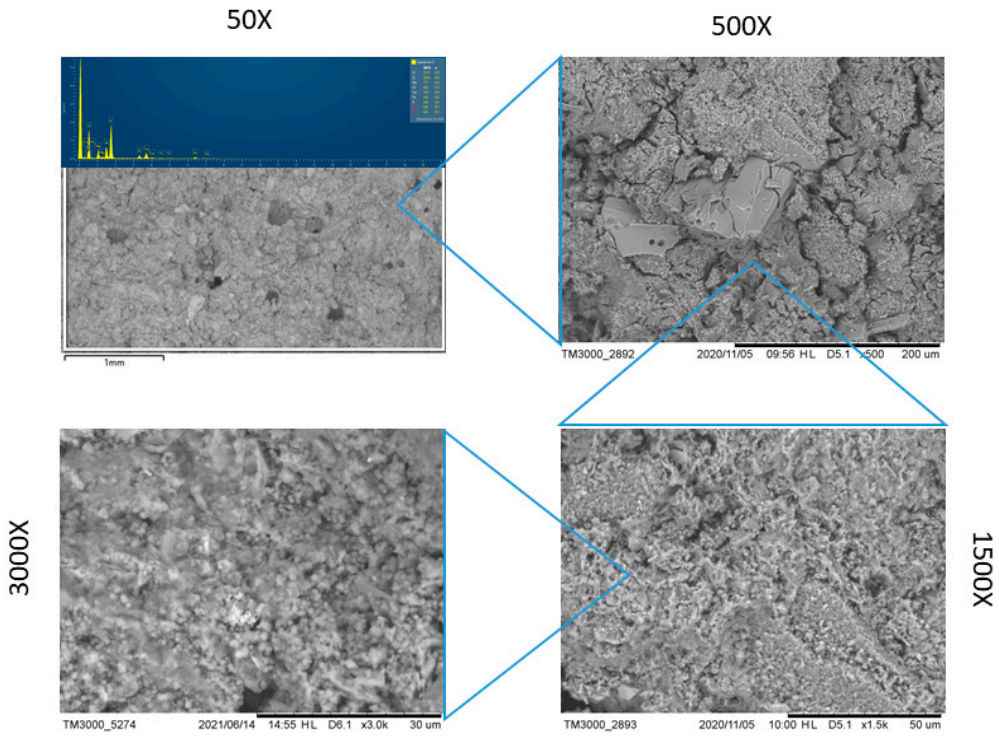

Figure 9. SEM images of the geopolymer structure at different magnifications: 500× (left) and 1500× (right). White circle (left): unreacted particles within the geopolymeric matrix.

The capillary absorption curves are reported in Figure 10. The data from the three tests indicated that the experimental method was performed with a high repeatability and that all the samples reached water saturation after 5–6 h. Moreover, the water absorption rate decreased for longer times, and this could be related to the increased water content inside the specimen and to the slow progressive participation of the less-accessible pores [32]. The average value of the capillary absorption coefficient is reported in Table 2, in which all the main physical properties of the CBW geopolymer are summarized. It is worth observing that the capillary absorption coefficient obtained from the CBW-based geopolymers was comparable to common values of the NHL mortars and, at the same time, lower than that of the typical solid clay brick (≈26) [33].

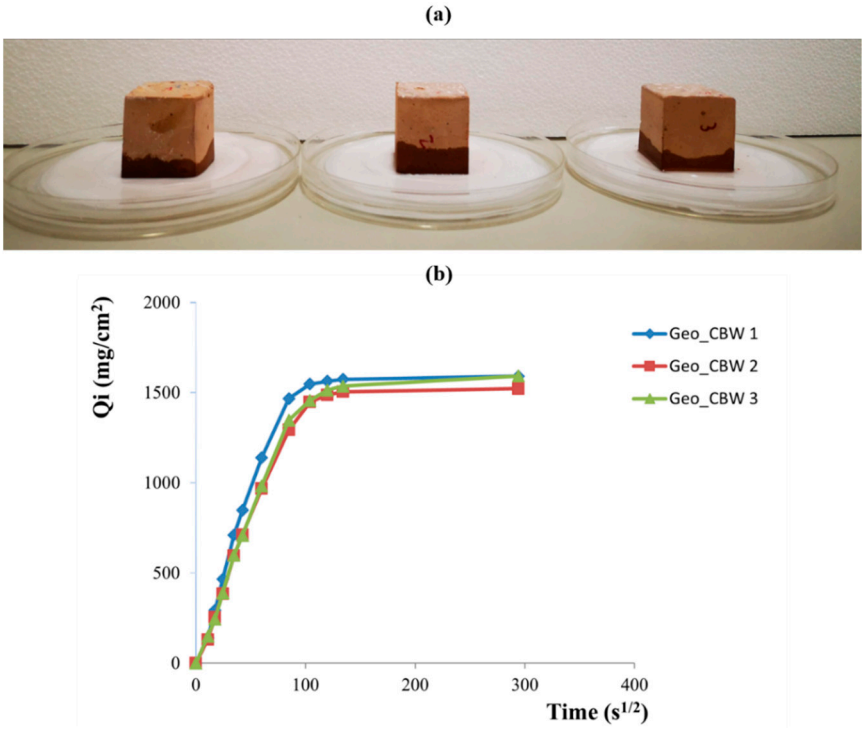

Figure 10. Capillarity test (**a**) and experimental results of the amount of water absorbed (Q) per surface unit in function of time (**b**) by the Geo_CBW samples.

Table 2. Physical properties of CBW geopolymer.

Apparent Density (g/cm^3)	Real Density (g/cm^3)	Open Porosity (%)
1.39 ± 0.02	2.56 ± 0.02	45.68 ± 0.37
Water absorption * (%)	**Water absorption (%)**	**CA (mg/cm^2 s$^{-1/2}$)**
16.65 ± 0.35	32.81 ± 0.67	18.24 ± 0.25

* values for immersion.

Regarding water absorption, it is worth noting that, as expected, the water absorption for immersion was almost half when compared to the value of the water absorption obtained by immersing the sample under a vacuum (see Table 2). The value of open porosity reported was about 46%, which further confirmed that the CBW-based geopolymeric samples produced were highly porous, as is clearly visible from the microstructural characterization (Figure 9).

The results obtained from the Shore D tests are shown in Table 3. The average value of the surface hardness (82.16) was reported, which is comparable to building plasters and gypsum composites [34,35].

Table 3. Shore D test results.

Sample	Point 1	Point 2	Point 3	Point 4	Point 5
Geo_CBW 1	85	76	85	86	90
	86	86	72	80	86
Geo_CBW 2	84	86	86	82	89
	76	85	73	81	80
Geo_CBW 3	85	80	75	86	86
	73	81	74	84	87

The flexural and compression tests provided a maximum resistance value of 2.85 ± 0.73 MPa and 5.34 ± 0.66 MPa, respectively. In order to grasp the possible applications of CBW, some of the results obtained from the Geo_CBW samples were compared to those found in the literature for several typologies of building materials that were comparable in terms of their mechanical behavior (see Table 4). In particular, different traditional building materials, such as lightweight gypsum and natural the hydraulic lime (NHL), were selected based on comparisons to different typologies of geopolymers and sustainable building materials produced from several kinds of waste.

Table 4. Comparison to results from other references.

Sample	Reference	Density (kg/m^3)	R_F (MPa)	R_C (MPa)
Geo_CBW		1390	2.85	5.34
Lightweight gypsum	[36]	910	1.52	2.17
Metakaolin-based foam	[37]	1000	0.14	4.62
Diatomite-based foam	[38]	423	0.63	1.49
NHL	[39]		2.15	5.55
NHL + 2% glass fibers	[39]	1590	2.41	3.62
NHL with plastic waste aggregate	[40]	1670	0.60	1.25
Dredged sediments geocomposite	[20]	/	/	1.90
Cement mortar with mixed recycled aggregate	[41]	1660	2.38	5.20
Gypsum + 1% polystyrene waste	[35]	970	2.89	5.64
Gypsum Plaster with ceramic waste from bricks	[42]	1180	2.80	5.40
Gypsum composites with glass waste	[34]	1270	2.93	6.01
Gypsum plaster with hemp fibers	[43]	/	2.50	/

From the data shown in Table 4, it is possible to deduce that the density and mechanical performance of the Geo_CBW samples were very similar to the gypsum samples produced from waste glass and ceramic waste made from brick similar to the CBW used in this paper.

4. Conclusions

Geopolymer manufacturing represents one of the most innovative ways to reuse different types of solid waste, especially when employed with aluminosilicate and clayey waste. In this paper, we investigated the potential of crushed brick waste as a raw material for the production of geopolymers. Preliminary chemical and mineralogical characterization were conducted on the CBW to evaluate if its chemical composition could be rendered suitable to undergo a geopolymerization process and to confirm its mainly clayey nature. Then, geopolymeric samples that were produced were widely characterized by means of chemical-physical, morphological, and mechanical laboratory testing. In particular, FTIR, XRD and SEM analyses confirmed that the geopolymeric reactions took place effectively. Evaluation of the physical and mechanical properties led to the conclusion that CBW can be successfully employed as a raw material in the production of geopolymer-based building precast components.

This approach of using secondary raw materials, derived from construction and demolition activities, in the production process of sustainable materials to reintroduce into the building market represents an intelligent and environmentally friendly solution to the significant environmental issues surrounding the disposal of this widely available waste.

Author Contributions: The paper is the result of a scientific work, carried out in collaboration by the authors. In particular, the contributions of each author are the following: Conceptualization, G.D., M.F. and M.d.R.M.; methodology, G.D., M.F., M.d.R.M. and B.L.; validation, M.F., M.d.R.M., D.C. and B.L.; formal analysis, G.D. and I.C.; investigation, G.D. and A.C.; resources, D.C., M.d.R.M., B.L. and F.I.; data curation, G.D., F.I. and I.C.; writing—original draft preparation, G.D., I.C. and A.C.; writing—review and editing, B.L., I.C., G.D. and M.F supervision, M.F., M.d.R.M., B.L. and D.C. All authors have read and agreed to the published version of the manuscript.

Funding: This research received no external funding.

Institutional Review Board Statement: Not applicable.

Informed Consent Statement: Not applicable.

Data Availability Statement: The data presented in this study are available on request from the corresponding author.

Acknowledgments: The authors thank the departments DiCMAPI and DICEA of the University of Naples Federico II, and the TEMA laboratory of the Polytechnic University of Madrid for their administrative and technical support.

Conflicts of Interest: The authors declare no conflict of interest.

References

1. Yuan, H.; Shen, L. Trend of the research on construction and demolition waste management. *Waste Manag.* **2011**, *31*, 670–679. [CrossRef]
2. Ghaffar, S.H.; Burman, M.; Braimah, N. Pathways to circular construction: An integrated management of construction and demolition waste for resource recovery. *J. Clean. Prod.* **2020**, *244*. [CrossRef]
3. Fischer, C.; Werge, M. *EU as a Recycling Society Present Recycling Levels of Municipal Waste and Construction & Demolition Waste in the EU*; European Topic Centre on Sustainable Consumption and Production: Copenhagen, Denmark, 2009.
4. Gálvez-Martos, J.L.; Styles, D.; Schoenberger, H.; Zeschmar-Lahl, B. Construction and demolition waste best management practice in Europe. *Resour. Conserv. Recycl.* **2018**, *136*, 166–178. [CrossRef]
5. Dos Reis, G.S.; Quattrone, M.; Ambrós, W.M.; Cazacliu, B.G.; Sampaio, C.H. Current applications of recycled aggregates from construction and demolition: A review. *Materials* **2021**, *14*, 1700. [CrossRef]
6. Capasso, I.; Liguori, B.; Ferone, C.; Caputo, D.; Cioffi, R. Strategies for the valorization of soil waste by geopolymer production: An overview. *J. Clean. Prod.* **2021**, *288*, 125646. [CrossRef]
7. Robayo-Salazar, R.A.; Valencia-Saavedra, W.; de Gutiérrez, R.M. Construction and demolition waste (Cdw) recycling—As both binder and aggregates—In alkali-activated materials: A novel re-use concept. *Sustainability* **2020**, *12*, 5775. [CrossRef]
8. Kvočka, D.; Lešek, A.; Knez, F.; Ducman, V.; Panizza, M.; Tsoutis, C.; Bernardi, A. Life cycle assessment of prefabricated geopolymeric façade cladding panels made from large fractions of recycled construction and demolition waste. *Materials* **2020**, *13*, 3931. [CrossRef]
9. De Brito, J.; Agrela, F.; Silva, R.V. Construction and demolition waste. In *New Trends in Eco-Efficient and Recycled Concrete*; Woodhead Publishing: Cambridge, UK, 2018; ISBN 9780081024805. [CrossRef]
10. Zhu, L.; Zhu, Z. Reuse of Clay Brick Waste in Mortar and Concrete. *Adv. Mater. Sci. Eng.* **2020**, *31*, 218–226. [CrossRef]
11. José, J.; Eras, C.; Sagastume, A.; Hernández, D.; Hens, L.; Vandecasteele, C. Improving the environmental performance of an earthwork project using cleaner production strategies. *J. Clean. Prod.* **2013**, *47*, 368–376. [CrossRef]
12. Coppola, L.; Bellezze, T.; Belli, A.; Bignozzi, M.C.; Bolzoni, F.; Brenna, A.; Cabrini, M.; Candamano, S.; Cappai, M.; Caputo, D.; et al. Binders alternative to Portland cement and waste management for sustainable construction—Part 2. *J. Appl. Biomater. Funct. Mater.* **2018**, *16*, 207–221. [CrossRef]
13. Youssef, N.; Rabenantoandro, A.Z.; Dakhli, Z.; Chapiseau, C.; Waendendries, F.; Hage Chehade, F.; Lafhaj, Z. Reuse of waste bricks: A new generation of geopolymer bricks. *SN Appl. Sci.* **2019**, *1*, 1252. [CrossRef]
14. Davidovits, J. Global Warming Impact on the Cement and Aggregates Industries. *World Res. Rev.* **1994**, *6*, 263–278.
15. Liguori, B.; Capasso, I.; De Pertis, M.; Ferone, C.; Cioffi, R. Geopolymerization Ability of Natural and Secondary Raw Materials by Solubility Test in Alkaline Media. *Environments* **2017**, *4*, 56. [CrossRef]
16. Ryu, G.S.; Lee, Y.B.; Koh, K.T.; Chung, Y.S. The mechanical properties of fly ash-based geopolymer concrete with alkaline activators. *Constr. Build. Mater.* **2013**, *47*, 409–418. [CrossRef]
17. Colangelo, F.; Cioffi, R.; Roviello, G.; Capasso, I.; Caputo, D.; Aprea, P.; Liguori, B.; Ferone, C. Thermal cycling stability of fly ash based geopolymer mortars. *Compos. Part B Eng.* **2017**, *129*, 11–17. [CrossRef]
18. Luhar, S.; Cheng, T.W.; Nicolaides, D.; Luhar, I.; Panias, D.; Sakkas, K. Valorisation of glass waste for development of Geopolymer composites—Mechanical properties and rheological characteristics: A review. *Constr. Build. Mater.* **2019**, *220*, 547–564. [CrossRef]
19. Kinnunen, P.; Ismailov, A.; Solismaa, S.; Sreenivasan, H.; Räisänen, M.L.; Levänen, E.; Illikainen, M. Recycling mine tailings in chemically bonded ceramics—A review. *J. Clean. Prod.* **2018**, *174*, 634–649. [CrossRef]
20. Lirer, S.; Liguori, B.; Capasso, I.; Flora, A.; Caputo, D. Mechanical and chemical properties of composite materials made of dredged sediments in a fly-ash based geopolymer. *J. Environ. Manag.* **2017**, *191*, 1–7. [CrossRef] [PubMed]
21. Capasso, I.; Lirer, S.; Flora, A.; Ferone, C.; Cioffi, R.; Caputo, D.; Liguori, B. Reuse of mining waste as aggregates in fly ash-based geopolymers. *J. Clean. Prod.* **2019**, *220*, 65–73. [CrossRef]
22. Ahmari, S.; Zhang, L. Production of eco-friendly bricks from copper mine tailings through geopolymerization. *Constr. Build. Mater.* **2012**, *29*, 323–331. [CrossRef]

23. UNI EN 15801:2010—Conservation of Cultural Property—Test Methods—Determination of Water Absorption by Capillarity 2010. Available online: http://store.uni.com/catalogo/uni-en-15801-2010 (accessed on 10 May 2021).
24. UNI 11060:2003—Beni Culturali—Materiali Lapidei Naturali ed Artificiali—Determinazione della Massa Volumica e Della Percentuale di Vuoti 2003. Available online: http://store.uni.com/catalogo/uni-11060-2003?josso_back_to=http://store.uni.com/josso-security-check.php&josso_cmd=login_optional&josso_partnerapp_host=store.uni.com (accessed on 10 May 2021).
25. EN 13279-2: 2014—Gypsum Binders and Gypsum Plasters—Part 2: Test Methods. 2014. Available online: https://standards.iteh.ai/catalog/standards/cen/7404ca86-7b9d-4584-945c-18b20c974ba2/en-13279-2-2014 (accessed on 10 May 2021).
26. Ferone, C.; Liguori, B.; Capasso, I.; Colangelo, F.; Cioffi, R.; Cappelletto, E.; Di Maggio, R. Thermally treated clay sediments as geopolymer source material. *Appl. Clay Sci.* **2015**, *107*, 195–204. [CrossRef]
27. Duxson, P.; Fernández-Jiménez, A.; Provis, J.L.; Lukey, G.C.; Palomo, A.; Van Deventer, J.S.J. Geopolymer technology: The current state of the art. *J. Mater. Sci.* **2007**, *42*, 2917–2933. [CrossRef]
28. Fernández-Jiménez, A.; Palomo, A.; Criado, M. Microstructure development of alkali-activated fly ash cement: A descriptive model. *Cem. Concr. Res.* **2005**, *35*, 1204–1209. [CrossRef]
29. Rees, C.A.; Provis, J.L.; Lukey, G.C.; Van Deventer, J.S.J. Attenuated Total Reflectance Fourier Transform Infrared Analysis of Fly Ash Geopolymer Gel Aging. *Langmuir* **2007**, *23*, 8170–8179. [CrossRef]
30. Swanepoel, J.C.; Strydom, C.A. Utilisation of fly ash in a geopolymeric material. *Appl. Geochem.* **2002**, *17*, 1143–1148. [CrossRef]
31. Verdolotti, L.; Iannace, S.; Lavorgna, M.; Lamanna, R. Geopolymerization reaction to consolidate incoherent pozzolanic soil. *J. Mater. Sci.* **2008**, *43*, 865–873. [CrossRef]
32. Alexandre Bogas, J.; Gomes, M.G.; Real, S. Capillary absorption of structural lightweight aggregate concrete. *Mater. Struct. Constr.* **2015**, *48*, 2869–2883. [CrossRef]
33. Karagiannis, N.; Karoglou, M.; Bakolas, A.; Moropoulou, A. Building Materials Capillary Rise Coefficient: Concepts, Determination and Parameters Involved. In *Building Pathology and Rehabilitation*; Springer: Singapore, 2016; Volume 6, pp. 27–44. [CrossRef]
34. Villoria Sáez, P.; del Río Merino, M.; Atanes Sánchez, E.; Santa Cruz Astorqui, J.; Porras-Amores, C. Viability of Gypsum Composites with Addition of Glass Waste for Applications in Construction. *J. Mater. Civ. Eng.* **2019**, *31*, 04018403. [CrossRef]
35. San-Antonio-González, A.; Del Río Merino, M.; Viñas Arrebola, C.; Villoria-Sáez, P. Lightweight material made with gypsum and extruded polystyrene waste with enhanced thermal behaviour. *Constr. Build. Mater.* **2015**, *93*, 57–63. [CrossRef]
36. Capasso, I.; Iucolano, F. Production of lightweight gypsum using a vegetal protein as foaming agent. *Mater. Struct. Constr.* **2020**, *53*, 35. [CrossRef]
37. Verdolotti, L.; Liguori, B.; Capasso, I.; Errico, A.; Caputo, D.; Lavorgna, M.; Iannace, S. Synergistic effect of vegetable protein and silicon addition on geopolymeric foams properties. *J. Mater. Sci.* **2014**, *50*, 2459–2466. [CrossRef]
38. Liguori, B.; Capasso, I.; Romeo, V.; D'Auria, M.; Lavorgna, M.; Caputo, D.; Iannace, S.; Verdolotti, L. Hybrid geopolymeric foams with diatomite addition: Effect on chemico-physical properties. *J. Cell. Plast.* **2017**, *53*, 525–536. [CrossRef]
39. Iucolano, F.; Liguori, B.; Colella, C. Fibre-reinforced lime-based mortars: A possible resource for ancient masonry restoration. *Constr. Build. Mater.* **2013**, *38*, 785–789. [CrossRef]
40. Verdolotti, L.; Iucolano, F.; Capasso, I.; Lavorgna, M.; Iannace, S.; Liguori, B. Recycling and recovery of PE-PP-PET-based fiber polymeric wastes as aggregate replacement in lightweight mortar: Evaluation of environmental friendly application. *Environ. Prog. Sustain. Energy* **2014**, *33*, 1445–1451. [CrossRef]
41. de Oliveira Andrade, J.J.; Possan, E.; Squiavon, J.Z.; Ortolan, T.L.P. Evaluation of mechanical properties and carbonation of mortars produced with construction and demolition waste. *Constr. Build. Mater.* **2018**, *161*, 70–83. [CrossRef]
42. San-Antonio-Gonzalez, A.; Santos Jimenez, R.; del Rio Merino, M.; Gonzalez Cortina, M.; Vinas Arrebola, C. Feasibility of Recycling CDW as Raw Material in Gypsum Composites. *Athens J. Technol. Eng.* **2015**, *2*, 149–160. [CrossRef]
43. Iucolano, F.; Boccarusso, L.; Langella, A. Hemp as eco-friendly substitute of glass fibres for gypsum reinforcement: Impact and flexural behaviour. *Compos. Part B Eng.* **2019**, *175*. [CrossRef]