

## Article

# Improving the Performance of Mortars Made from Recycled Aggregates by the Addition of Zeolitised Cineritic Tuff

Domingo A. Martín <sup>1,2</sup>, Jorge L. Costafreda <sup>1,\*</sup>, Jorge L. Costafreda, Jr. <sup>3</sup> and Leticia Presa <sup>1</sup>

<sup>1</sup> Escuela Técnica Superior de Ingenieros de Minas y Energía, Universidad Politécnica de Madrid C/Ríos Rosas, 21, 28003 Madrid, Spain; domingoalfonso.martin@upm.es (D.A.M.); leticia.presa.madrigal@upm.es (L.P.)

<sup>2</sup> Laboratorio Oficial para Ensayos de Materiales de Construcción (LOEMCO), C/Eric Kandell, 1, 28906 Getafe, Spain

<sup>3</sup> Empresa de Proyectos Vértice, Departamento de Estructuras, Avenida XX Aniversario, Reparto Piedra Blanca, Holguín 80949, Cuba; costafreda1992@gmail.com

\* Correspondence: jorgeluis.costafreda@upm.es; Tel.: +34-609642209

**Abstract:** Metropolitan construction and demolition waste (CDW) is currently an important source of recycled materials that, despite having completed their useful life cycle, can be reincorporated into the circular economy process (CEP); however, the recycling process is very selective, and waste material is not always fully satisfactory due to the intrinsic nature of the waste. This work aims to demonstrate and establish how to increase the effectiveness of the construction and demolition waste in more resistant mortars, by mixing it with zeolitised cinerite tuff (ZCT) at varying normalised proportions. To attain the objectives of this research, a series of tests were done: First, a chemical, physical and mineralogical characterisation of the CDW and the ZCT through XRF, XRD, SEM and granulometric methods. Second, a technological test was made to determine the mechanical strength at 7, 28 and 90 days of specimens made with Portland cement (PC) and mixtures of PC/CDW, PC/ZCT, and PC/CDW-ZCT. The results obtained through the characterisation methods showed that the sample of construction and demolition waste consisted of the main phase made of portlandite and tobermorite, and by a secondary phase consisting of quartz, ettringite and calcite; whereas the ZCT has a main phase of mordenite and a secondary phase of smectite (montmorillonite), amorphous materials consisting of devitrified volcanic glass, quartz and plagioclase. Mechanical strength tests established that specimens made with PC/CDW mixtures have very discreet compressive strength values up to 44 MPa at 90 days, whereas specimens made with PC/ZCT mixtures achieved a remarkably high mechanical strength consisting of 68.5 MPa. However, the most interesting conclusion in this research is the good result obtained in mechanical strength of the specimens made up of mixtures of PC/CDW-ZCT, which increased from 52.5 to 62 MPa at 90 days of curing; this fact establishes the positive influence of ZCT on waste in the mortar mixtures, which permits the authors to establish that the objective of the work has been fulfilled. Finally, it can be argued that the results obtained in this research could contribute to more effective use of construction and demolition waste in metropolitan areas.

**Keywords:** construction and demolition waste; zeolitised cinerite tuff; cement; mechanical strength



**Citation:** Martín, D.A.; Costafreda, J.L.; Presa, L. Improving the Performance of Mortars Made from Recycled Aggregates by the Addition of Zeolitised Cineritic Tuff. *Crystals* **2022**, *12*, 77. <https://doi.org/10.3390/crust12010077>

Academic Editors: Edyta Pawluczuk, Iwona Skoczko and Enrique Fernandez Ledesma

Received: 23 December 2021

Accepted: 4 January 2022

Published: 6 January 2022

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



**Copyright:** © 2022 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

An environmental emergency is pressing us today. The growing geopolitical conflicts of the world's major economic blocs are rooted in the availability and management of natural resources. The idea of the possible depletion of resources in the short term has awakened the need to look for new alternatives, such as the reuse of materials from old buildings, which could be a definitive alternative, since large volumes of construction and demolition waste are accumulated every year in the great metropolises of the world, whose degradation by weathering agents seriously damages soils, aquifers, air quality,

among others. Pacheco-Torgal et al. [1] mention that the European construction sector produces about 890 million tonnes of waste per year, and point out that the amended Waste Framework Directive aimed to recycle 70% of construction and demolition waste by 2020; however, with the exception of a few Member States, only 50% is recycled.

The use of construction and demolition waste (CDW) in the improvement of the quality of mortars is an increasingly common practice nowadays, as is seen in the great amount of research done and published in the last decade, which results in a great deal of knowledge achieved in this area. Following are some relevant works regarding construction and demolition waste. Jesus et al. [2] perfected mortars made with construction and demolition waste ground to 0.149 mm and then mixed in the cement coatings, where the natural aggregate (NA) was replaced by CDW, managing to raise the mechanical strength by substituting 20% of NA with CDW. Azevedo et al. [3] replaced natural sand with these wastes in mortars in 25, 50 and 100% and established that 25% substitution is the most suitable to obtain better compaction and the highest mechanical strengths. Silva et al. [4] evaluated the performance of mortars made from residues consisting of ceramic material, bricks and clay tiles, with satisfactory results in substitutions of up to 20% of natural sand by construction and demolition waste. Whereas Oliveira et al. [5] and Infante et al. [6] obtained mortars mixed with CDW with a greater CO<sub>2</sub> absorption potential capable of capturing up to 170 g of CO<sub>2</sub>/m<sup>2</sup>. Fernandez et al. [7] developed a fine aggregate based on construction and demolition waste with a particle size of 4 mm and achieved greater effectiveness with a 50% formulation in mortar mixes. Other researchers such as Saiz et al. [8] and Katz and Kulisch [9] consider that recycled materials are more efficient when several recycled materials are mixed together. Duan et al. [10], Duan et al. [11], Jesus et al. [12], Colangelo and Cioffi [13] and Ferreira et al. [14] have managed to raise the mechanical strength of mortars made from ultrafine powder from bricks and calcinated clays, with proportions of 20 and 30%; they have also achieved good results by replacing part of the Portland cement, fly ash and natural sand in the concrete with recycled powders. A similar result has been obtained by De Rossi et al. [15] in their investigations made with various sizes of fly ash particles to monitor the behaviour of geopolymers in a fresh and hardened state, with a total substitution of normalised sand. Sales and Rodriguez [16] have obtained high mechanical strengths in mortars and concretes made with a mixture of sludge from water treatment plants of construction and demolition waste, which also improved some parameters such as elasticity, water absorption and tensile strength. Del Rio-Merino et al. [17] have conducted an extensive review of the use of recycled gypsum in the manufacture of buildings, considering this practice as a corrective measure of environmental impact. In the same line, Antunes et al. [18] have proposed the use of recycled gypsum in the manufacture of mortars. Robayo-Salazar et al. [19] put forth a mixture of concrete, ceramic, masonry and mortar residues activated by alkalis and mixed with only 10% Portland cement (PC), obtaining mechanical strengths of up to 43.9 MPa at 28 days of curing. Wu et al. [20] have conducted an excellent state-of-the-art critical review of the use of construction and demolition waste since 1990; they, therefore, predict that this knowledge could be used well in important fields such as material engineering, industrial ecology, management science and architecture, thus establishing a relationship model of these subject matters with the environment. Stefanidou et al. [21] have put forth a novel system consisting of the manufacture of sand based on recycled lime rich in SiO<sub>2</sub> and Al<sub>2</sub>O<sub>3</sub>, to be used in mortars for the restoration of historic buildings, obtaining promising results. Asensio et al. [22] studied the properties of newly designed eco/efficient pozzolanic cement, composed of residues from industrial processes and by-products based on calcined clay. According to the results obtained, these types of cement showed physical, chemical and mechanical properties in accordance with standard parameters and limits. Shahmansouri et al. [23] have improved the mechanical and durability properties of concrete by using natural zeolites, showing that the addition of zeolite in the mix (7.5–10%) significantly increased the mechanical strength, while additions of 20% increased the electrical resistivity of the mixes at higher ages. Recently, Shahmansouri et al. [24] have conducted experimental studies to investigate

the effect of sodium hydroxide (NaOH) concentration and substitution of GGBFS with pozzolans of zeolitic nature and silica fume on the mechanical properties of geopolymers concrete; the results showed that the presence of natural zeolite in the mix improves the compressive, flexural and tensile strengths of geopolymers concrete with 10% substitution.

Among the main objectives of this work is to monitor the mechanical behaviour of a series of specimens designed with different proportions of mixtures, in which the contents of CDW vary in relation to NA, until obtaining a final mixture that totally replaces NA with CDW. In the formulation of these mixtures, standardised portions of ZCT are also introduced, which partially replace Portland cement (PC). The final goal is to obtain an ideal mixture that significantly saves on NA and PC and at the same time has high mechanical strength. The results provided by this research could meet several other expectations, such as (a) the conservation of industrial rock and mineral deposits, which have been indiscriminately exploited for decades and could lead to the depletion of reserves; (b) the effective management of CDWs as a culture of European countries in the efficient use of the large volumes of waste produced per year; (c) the circular economy, based on the reuse of materials that have completed its life cycle and their valorisation, and (d) environmental preservation, as the higher the consumption of CDW, the lower the impact of mining on natural deposits and the shorter the exposure time of these inert materials in the environment.

## 2. Materials and Methods

### 2.1. Materials

A global sample of recycled aggregate from construction and demolition waste from the Community of Madrid (Spain) was chosen for this research. The sample was carefully selected from the well-preserved concrete waste storage sites. Another sample of zeolitised cinerite tuff (ZCT) from the volcano-sedimentary formations of the south of the Iberian Peninsula was chosen for its physical, chemical, mineralogical and mechanical characterisation. In this research we worked with Portland cement (PC) type I, strength class CEM I 42.5 R with characteristics, properties and conformity defined according to the Standard UNE-EN 197-1:2011 [25].

A normalised sand (NS) coded as CEN-NORMSAND DIN EN 196-1 was used as a fine natural aggregate (NA) in this research, to determine the mechanical strengths of Portland cement and mortars with different mixture formulations. The data on the particle distribution, determined by the Standard UNE-EN 933-1:2012 [26] for the NA and CDW samples in this research, are shown in Table 1.

**Table 1.** Particle size distribution of CDW and natural aggregate (NA) particles calculated in this research.

Natural Aggregate (NA)		Construction and Demolition Waste (CDW)	
Size (mm)	Retained Mass (%)	Size (mm)	Retained Mass (%)
2.0	0	2.0	0
1.0	30.9	1.0	34.6
0.5	34.4	0.5	30.2
0.25	11.3	0.25	17.7
0.125	20.4	0.125	9.0
0.063	2.9	0.063	2.4
-0.063	0.1	-0.063	6.1
Total	100	total	100

## 2.2. Methods

### 2.2.1. X-ray Diffraction (XRD)

X-ray diffraction analysis was carried out to determine the dominant mineral phases in both the CDW sample and the ZCT sample. This analysis was performed following the standard procedure of the crystal powder method (PTE-RX-004). Measurements were made with XPERT-PRO MPD equipment, PANalytical (Malvern, UK) brand. This equipment has a copper tube (45 kV, 40 mA) and a graphite monochromator with an automatic opening. The X'Pert Data Collector 5.1 (5.1.0.156) of PANalytical was used to collect the data. On the other hand, the HighScore software version 3.0.4 (PANalytical) from the Escuela Técnica Superior de Ingenieros de Minas y Energía (Universidad Politécnica de Madrid, Spain) and the PDF-2 (ICDD) and CODJanuary2012 databases were used, to further analyse and interpret the acquired results.

### 2.2.2. Scanning Electron Microscopy (SEM)

A Hitachi S-570 scanning electron microscope from the Laboratorio Centralizado of the Escuela Técnica Superior de Ingenieros de Minas y Energía (Universidad Politécnica de Madrid, Spain) was used in the investigation of the CDW and ZCT samples, respectively. The equipment consists of a Kevex 1728 analyser, a BIORAD-Polaron Division Carbon evaporation power supply, as well as by an SEM-Polaron type coating system. Other components that make up this equipment are a silicon semiconductor detector tube with Li alloy, a liquid nitrogen reservoir, a filament chamber, an electronic cannon, a sample introduction-extraction system in the high vacuum chamber, a system to vary the angles of the position of the sample, an image visualisation module, as well as two software: Winshell and Printerface, which manages the data obtained, including taking microphotographs. The following procedure was done to carry out the analysis: samples were reduced to between 0.2–0.5 cm; graphitisation, placed in the sample holder and introduced into the high vacuum chamber of the electron microscope.

### 2.2.3. X-ray Fluorescence (XRF)

The analysis of the CDW, ZCT, NA and PC samples were done using the X-ray fluorescence test (XRF), to determine their chemical composition. A Phillips fluorescence device model PW-1404 (Madrid, Spain) was used in this analysis. This equipment is made up of a collimator designed to decrease the value of the angle of divergence of the X-rays, as well as to mitigate deviations and reinforce the beam. The radiation intensity value on the samples was 10–100 kV. A monochromator was used to isolate the radiation and obtain a good wavelength. To carry out this analysis, 8 g of each sample were crushed up to 74 µm, were mixed in 1.5 mL of a solution made of 250 cc of acetone and 12.5 g of plastic to give the sample binding properties and avoid ruptures during pressing. The pressing was carried out with the help of the Herzog press. The Heraeus muffle at 1000 °C was used to determine loss on ignition (LOI).

### 2.2.4. Tests to Determine the Specific Area, Sand Equivalent, Water Absorption, Organic Matter Content and Friability Coefficient of the CDW

The CDW sample was also investigated by technological assays, such as specific surface area (SS), sand equivalent (SE) and water absorption (WA); to carry out these studies, the procedures in the Standards UNE-EN 933-9:2010 [27], UNE-EN 933-8:2012 [28] and UNE-EN 1097-6:2000 [29] were rigorously followed.

To determine the specific surface (SS) a sample of CDW with a particle size of less than 2 mm was used. A portion of methylene blue (MB) was used in this trial. The amount of MB needed to adequately cover 1 kg of sample was determined by Equation (1):

$$MB = \frac{V_1 - V_2}{M_1} \times 10 \quad (1)$$

whereby:

$V_1$ : volume of the added solution

$V_2$ : volume of the solution absorbed by the particles

$M_1$ : dry sample mass

The sand equivalent (SE) was calculated using Equation (2):

$$SE = \frac{h_2}{h_1} \times 100 \quad (2)$$

whereby:

$h_1$ : height of the upper level of flocculate

$h_2$ : penetration height of the weighted baton (sediment height)

To determine water absorption (WA) by immersion, the water temperature was set at 21 °C. The mass of the CDW sample was determined after 24, 48 and 72 h after immersion, with the help of Equation (3):

$$WA = \frac{Me_{48} - Me_{24}}{Me_{24}} \times 100 \quad (3)$$

whereby:

$Me_{48}$ ,  $Me_{24}$ : sample mass at different periods of immersion

After the periods of immersion alluded to above, the sample was dried in a muffle at a temperature of 105 °C ± 5 °C. The weight of the dry sample (Wds) was determined by Equation (4):

$$Wds = \frac{Ms_{48} - Ms_{24}}{Ms_{24}} \times 100 \quad (4)$$

whereby:

$Ms_{48}$ ,  $Ms_{24}$ : dry sample weight at different periods

The content of organic matter (OMC) was determined according to Standard UNE-EN 1744-1:2010 [30]. The CDW sample was dried on a stove at 50 °C for 24 h. The sample was then mixed in a 3% sodium hydroxide solution, then vigorously stirred and left to stand for 24 h.

The friability coefficient (FC) was determined given the uneven nature of CDW mainly regarding hardness, coherence and mechanical strength. The development of this test was based on the precepts of the Standard UNE 83-115-89 [31]. A sample ( $M_1$ ) of CDW weighing 500 g was dried on a stove at 100 °C for 24 h. It was then introduced into the Micro/Deval mill along with 2.5 L of distilled water and 1.5 kg of steel balls. The grinding process lasted 15 min. The material was then extracted and sieved in the 5 mm, 0.32 mm and 0.050 mm sieves. Subsequently, the material was washed and striped of the finest fraction. The material retained in the 0.050 mm sieve was dried for 24 h, after which it was weighed ( $M_2$ ).

## 2.2.5. Mechanical Strength Test at 7, 28 and 90 Days

To determine the mechanical strength to compression at 7, 28 and 90 days, eight specimens were made, taking as a reference the guide provided by the Standard UNE-EN 196-1:2018 [32]. Before formulating the different mixing proportions, the ZCT sample was ground and sieved at 63 µm. To carry out this test a project of mixtures of materials with different natures was made as seen in Table 2.

The PCS specimen was made with the components of an ordinary mortar, in which there is 100% natural aggregate (NA), 100% Portland cement (PC) and 225 g of distilled water. This sample does not include other aggregates and is used in this research as a reference specimen. In the PC/ZCT sample part of the PC is replaced by ZCT, in a ratio of 75:25. In the samples PC/CDW-ZCT-01 to 04, new equivalent quantities of materials were formulated as indicated in Table 2, by means of which NA for CDW substitutions were made in 75, 50, 25 and 0%, while the PC/ZCT ratios remained fixed. In this case, the

amount of water was increased by 25 g, given the absorption capacity of the CDW and the ZCT. Finally, a sample composed exclusively of PC/CDW was prepared to determine the ability of the CDW to generate mechanical strength in mortars.

**Table 2.** Formulation of mortar mixtures made with different proportions of PC, NS, CDW, ZCT and DW, in accordance with the procedures of the Standard UNE-EN 196-1:2018 [32].

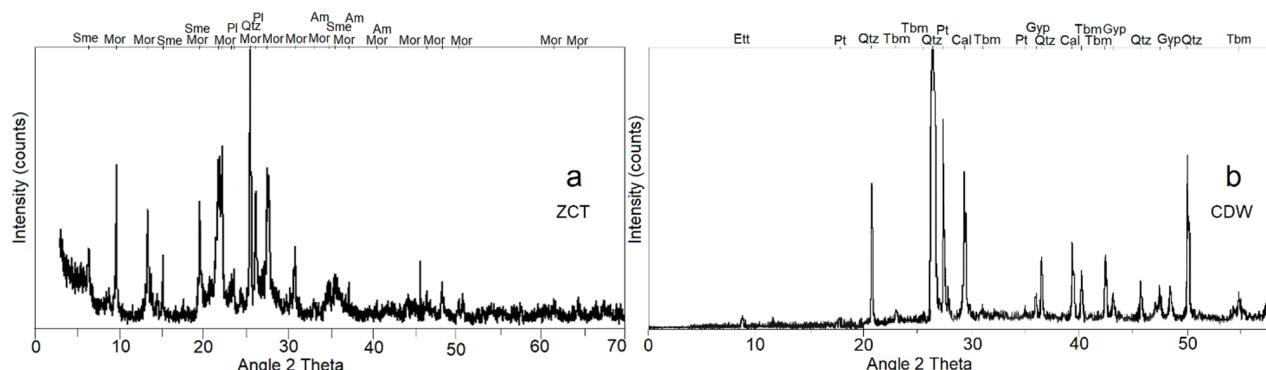
Nomenclature of Specimens	Mortar Components				
	NA <sup>1</sup> (%)	CDW <sup>2</sup> (%)	ZCT <sup>3</sup> (%)	PC <sup>4</sup> (%)	DW <sup>5</sup> (g)
PCS <sup>6</sup>	100	0	0	100	225
PC/ZCT <sup>7</sup>	100	0	25	75	225
PC/CDW-ZCT-01 <sup>8</sup>	75	25	25	75	250
PC/CDW-ZCT-02	50	50	25	75	250
PC/CDW-ZCT-03	25	75	25	75	250
PC/CDW-ZCT-04	0	100	25	75	250
PC/CDW <sup>9</sup>	0	100	0	75	225

<sup>1</sup> Natural aggregate (sand); <sup>2</sup> Construction and demolition residue; <sup>3</sup> Zeolitised cineritic tuff; <sup>4</sup> Portland cement; <sup>5</sup> Distilled water; <sup>6</sup> Portland cement specimen used as reference; <sup>7</sup> Portland cement and zeolitised cineritic tuff specimen; <sup>8</sup> Portland cement specimens with construction and demolition waste and zeolitised cineritic tuff; <sup>9</sup> Portland cement specimen with construction and demolition waste.

### 3. Results

#### 3.1. X-ray Diffraction (XRD)

The results of the X-ray diffraction analysis (XRD) revealed the presence of different phases in the mineral composition of the samples investigated, both from the ZCT and the CDW (Figure 1a,b). The mineral phases detected in the ZCT sample (Figure 1a) are complex in nature, and are as follows: mordenite (Mor), smectite (Sme) of montmorillonite variety, amorphous materials (Am) in the form of devitrified volcanic glass (DVG), quartz (Qtz) and plagioclase (Pl). The dominant phases, in order of hierarchy, are mordenite and smectite.



Mor: Mordenite; Sme: Smectite (Montmorillonite); Am: amorphous (Volcanic glass); Pl: Plagioclase; Qtz: Quartz; Pt: Portlandite; Tbm: Tobermorite; Ett: Ettringite; Cal: Calcite; Gyp: Gypsum

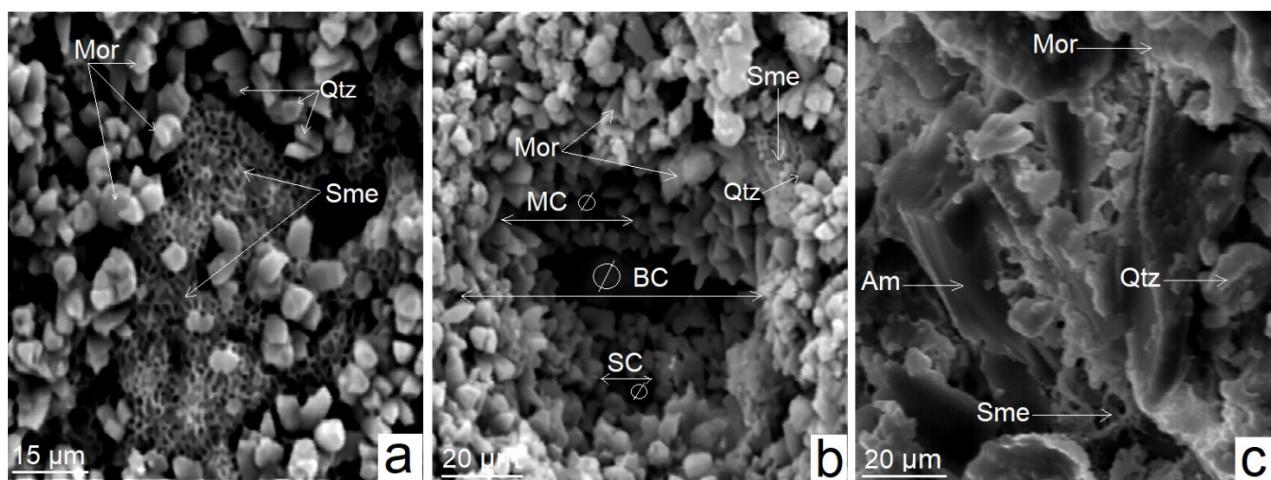
**Figure 1.** X-ray diffraction patterns of researched samples: (a) sample ZCT, (b) sample CDW.

In the CDW sample (Figure 1b) several mineral phases consisting of portlandite (Pt), tobermorite (Tbm), ettringite (Ett), quartz (Qtz), calcite (Cal) and gypsum (Gyp) were detected. The presence of tobermorite and portlandite phases infers that the recycled aggregate comes from ancient mixtures where a cement that was normally cured was present. The presence of the ettringite phase (Ett) indicates that the original cement contained tricalcium aluminate in its composition [33].

According to the phases mentioned above, it is evident that the ZCT, originated in a volcanic environment of pyroclastic nature, as indicated by the cinerite and vitreous nature of the tuff host of the zeolitic and clay mineralisation, where the hydrothermal alterations have caused the most abundant mineral species, such as those mentioned above; this deduction is remarkably close to what was previously established by Costafreda [34].

### 3.2. Scanning Electron Microscopy (SEM)

The microphotographs in Figure 2a–c show the presence of various mineral species present in the sample of ZCT, such as mordenite, smectite, quartz and volcanic glass. A predominance of mordenite (Mor) with a high degree of crystallinity can be observed in all cases. Figure 2a shows the mordenite-smectite paragenesis; note also the typical “honeycomb” texture that characterises smectites (Sme) and persists in Figure 2b. The coexistence of both mineral phases seems to reinforce their syngenetic origin from the devitrification of volcanic glass, as can be seen in Figure 2c. Costafreda [34] and later Presa et al. [35] have described this allochemical process on several occasions, and this fact significantly reinforces what is stated in this research.



Mor: Mordenite; Sme: Smectite (montmorillonite); Qtz: Quartz; Am: amorphous (volcanic glass)  
BC: Big cavity; MC: Middle cavity; SC: Small cavity

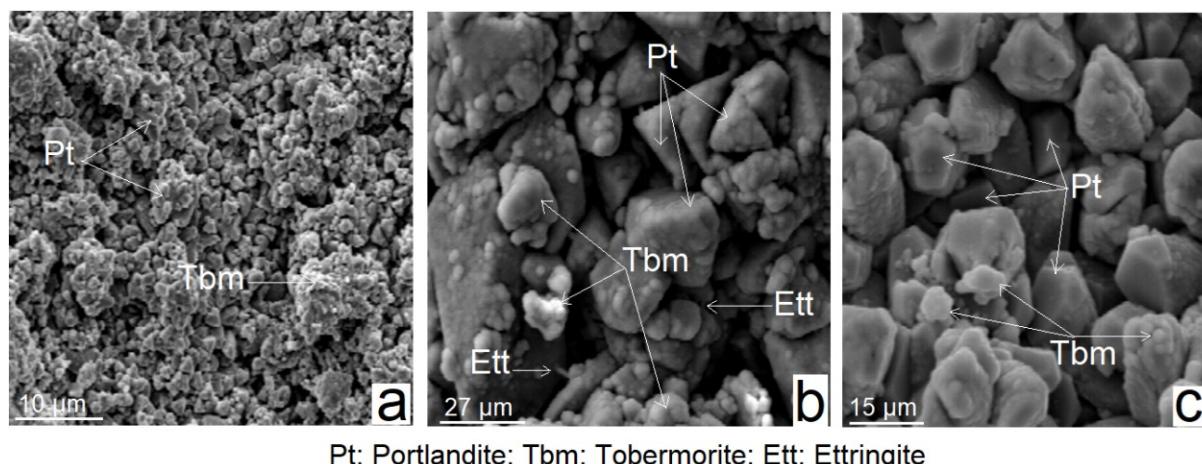
**Figure 2.** Microphotographs (a–c) of the ZCT sample, obtained through scanning electron microscopy (SEM).

The typical porous structure of mordenite, as with the rest of the zeolite family [36], is clearly seen in Figure 2b, where several cavities of different sizes can be seen. In this research, a specific classification of these openings has been made as follows: big cavity (BC), medium diameter cavity (MC) and small diameter cavity (SC). The presence of cavities, channels and pores in wide varieties of zeolites has been mentioned in many works, among which are highlighted the conclusions provided by Korkuna et al. [37], Nilchi et al. [38] and Flanigen [39].

The minerals phases detected by SEM are practically the same as those determined by XRD, therefore, both analyses have been suitable for the purposes of this research and reinforce each other.

Figure 3a–c shows the microphotographs obtained by scanning electron microscopy (SEM) of the CDW sample. Basically, the composition of this sample consists of portlandite (Pt), tobermorite (Tbm) and ettringite (Ett), where the predominant phases are portlandite and tobermorite. Given the development of these phases, it can be deduced that the CDW used in this research is of good quality from a physical, chemical and mineral point of view; the development of the tobermorite phase confirms this fact [40]. The presence of portlandite is favourable for hydraulic reactions of the pozzolanic interfaces consisting of zeolite and other pozzolans, since it tends to react with the Pt to form more stable reaction

products, such as Tbm, which provides physical and chemical strength to the mortar and concrete. Rosell et al. [41] have largely reflected on this fact in their studies on the kinetics of pozzolanic reactions, which seems to coherently confirm the deductions made in this investigation.



**Figure 3.** Microphotographs (a–c) of the CDW sample studied in this work, made with the scanning electron microscope.

### 3.3. X-ray Fluorescence (XRF)

Table 3 shows the results of the chemical composition of the samples investigated by X-ray fluorescence (XRF). The first aspect to highlight is the high content of  $\text{SiO}_2$  in all samples, the highest value is the natural aggregate (NA), followed by the zeolitised cinerite tuff (ZCT), while the smallest amount corresponds to the Portland cement sample (PC). In the specific case of construction and demolition waste (CDW), the presence of  $\text{CaO}$  was determined at 22.1%, the highest value after PC; however, the percentage of  $\text{Al}_2\text{O}_3$  is rather low, as is also the case with  $\text{SO}_3$ . The NA make-up is practically monomineral, while ZCT has a more complex and varied chemical composition, which reflects the presence of several mineralogical varieties. The highest values of loss on ignition (LOI) corresponds to the CDW and ZCT samples, respectively, which is very low for the rest of the samples investigated.

**Table 3.** Chemical composition of the samples investigated was determined by X-ray fluorescence (XRF).

Sample	Anhydrous Compounds in % Mass								
	$\text{SiO}_2$	$\text{CaO}$	$\text{Fe}_2\text{O}_3$	$\text{Al}_2\text{O}_3$	$\text{SO}_3$	$\text{Na}_2\text{O}$	$\text{MgO}$	$\text{K}_2\text{O}$	LOI *
CDW	54.17	22.1	1.82	5.86	1.31	-	0.298	1.67	12.22
NA	94.25	0.3	0.88	2.24	0.06	0.059	0.06	0.86	0.48
ZCT	68.5	1.20	1.46	11.97	-	2.93	1.32	1.42	12.73
PC	18.76	64.5	3.81	5.24	4	0.082	1.11	1.27	2.13

\* LOI: Loss on ignition.

The high silica content of the CDW may be due to the presence of phases of natural aggregates from the primary stage of concrete processing, as well as the silicate phases of neoformation, such as tobermorite (Tbm), mentioned before in Sections 3.1 and 3.2.

The absence of  $\text{Na}_2\text{O}$  and the low content of  $\text{K}_2\text{O}$  is a positive indicator that highlights the good quality of the researched CDW, mainly since the low presence of alkaline compounds could prevent their harmful influence on the structural stability of the specimens studied [40]. On the other hand, the high LOI values, calculated for the CDW and ZCT samples, point to these materials as being chemically reactive [41].

By paying close attention to the SiO<sub>2</sub> and CaO contents of PC and CDW (Table 3) it shows that the latter has a CaO deficit of approximately 37% compared to the PC; however, the CDW exceeds the PC by 34% more of SiO<sub>2</sub>, thus establishing an inversely proportional relationship in which the chemical composition of the PC and the CDW seems to compensate each other. According to the information in Table 3 on the chemical composition of the CDW, phases of calcium silicates, as well as calcium aluminates and ferroaluminates, could be formed in the mortar, characterised by a high hydraulic reactivity [42].

#### 3.4. Specific Area (SS), Sand Equivalent (SE), Water Absorption (WA), Organic Matter Content (OMC) and Friability Coefficient (FC) of the CDW

The results of technological tests, such as specific area (SS) [27], sand equivalent (SE) [28], water absorption (WA) [29], organic matter content (OMC) [30] and friability coefficient (FC) [31] of the CDW are set out in Table 4.

**Table 4.** Results of the technological quality tests of the CDW were investigated in this work.

Construction and Demolition Waste (CDW)	
Tests	Results
Specific surface (SS) (m <sup>2</sup> /g)	2.88
Sand equivalent (SE) (%)	60.8
Water absorption by immersion WA (%)	4.64
Organic matter content (OMC)	No organic matter
Friability coefficient (FC) (%)	18.60

According to the results presented in Table 4, the value of the specific surface of the CDW is low compared to the ZCT. Costafreda [34] calculated a specific surface for mordenite equivalent to 66 m<sup>2</sup>/g, so it follows that this difference could be compensated by the high hydraulic reactivity of the ZCT, inferred by the extremely large specific surface of mordenite, as mentioned above. The value of the sand equivalent (SE) is considered high and therefore adequate for the objectives of this research. This value (60.8%) indicates that the greater its tendency to increase, the lower the content of fine particles, which positively influences the increase in mechanical strength of the mortar. Comparatively, the water absorption capacity of the CDW is relatively low, although not negligible with respect to ZCT in the formulation of the mixtures, which requires an increase in the proportion of water to achieve more workable mortars. This research establishes that the amount of additional water should be set at 25 g.

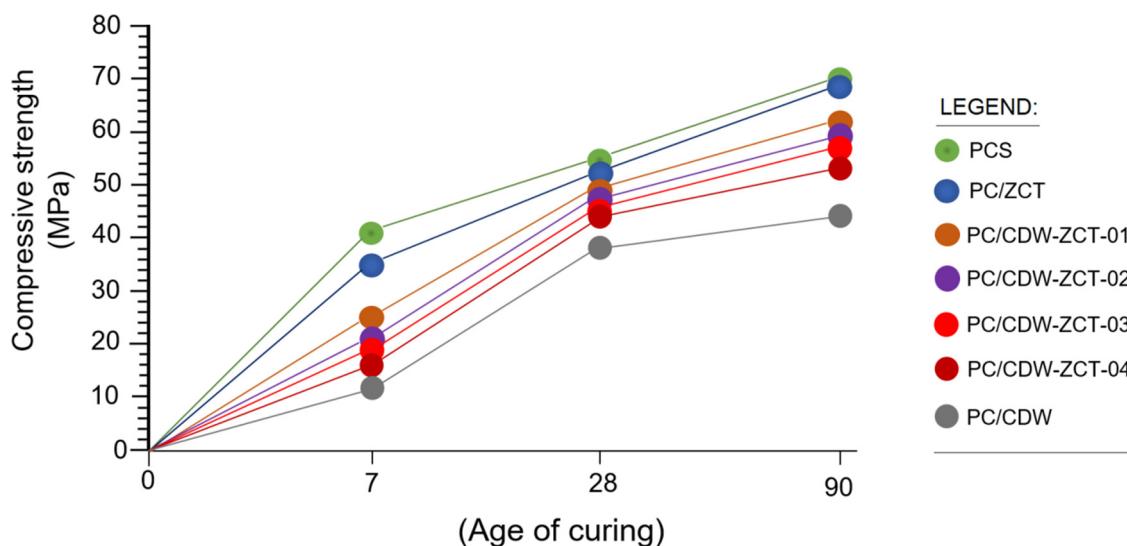
The value of the friability coefficient (18.60%) indicates that the CDW analysed is adequate, in relation to the specifications of Standard UNE 83-115-89 [31], which governs the test procedure; therefore, it is suitable for the objectives.

#### 3.5. Mechanical Strength to Compression Test at 7, 28 and 90 Days

Figure 4 shows the results of the mechanical strength to compression obtained from the assessment of 7 specimens at the ages of 7, 28 and 90 days. During this time period, all samples showed increasing but uneven hydraulic reactivity. The order from highest to lowest reactivity of each specimen is as follows: PCS, PC/ZCT, PC/CDW-ZCT (01 to 04) and PC/CDW.

The PCS specimen, which represents the reference specimen, has a short setting time and the greatest capacity to acquire great resistances from the initial periods of setting [41]; according to Figure 4, the compressive strength values of the PCS are increased substantially from 7 to 90 days, compared to the remaining specimens. The specimen made of PC/ZCT has the second-highest value of compressive strength, although it replaces the PC by 25%, which confirms its good properties as a pozzolan; this fact is highlighted, fundamentally, in the curing periods between 28 and 90 days, in which the difference between the values of

mechanical strength values of the PC/ZCT with respect to the reference specimen (PCS) decreases drastically. Sometimes a pozzolan, such as the ZCT used in this work, exceeds the value of the mechanical strength of Portland cement (PC) after 28 days, as has already been demonstrated several times by Costafreda [34], who has achieved compressive strengths in specimens with zeolites of 52, 70 and 76 MPa at 28, 90 and 365 days, respectively, surpassing in all cases the strengths of Portland cement.

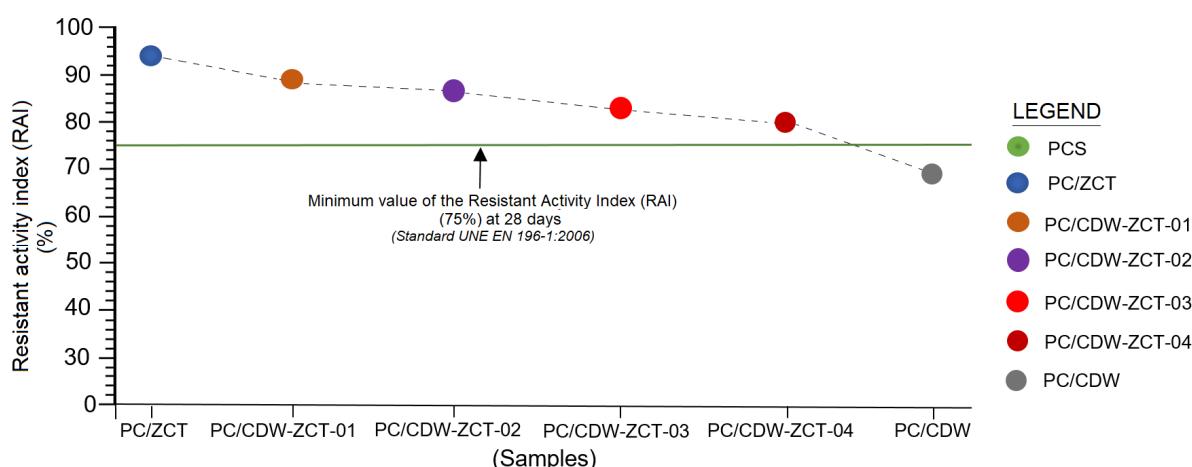


**Figure 4.** Mechanical strength to compression determined for specimens made with PCS, PC/ZCT, PC/CDW-ZCT and PC/CDW at 7, 28 and 90 days of curing.

In the PC/CDW-ZCT specimen series (01 to 04) maximum values of mechanical strength to compression are observed in cases where the dosages include 25% CDW, 25% ZCT, 75% PC and 75% NA. These values decrease significantly to the extent that the percentages of CDW in the specimens increase and those of NA decrease; however, the mechanical strength remains adequate in all cases. A very different situation occurs when analysing the PC/CDW specimen, which has the lowest values of mechanical compressive strength throughout the entire investigation period, similar to that proposed by Da Silva and De Oliveira [43]. In the PC/CDW sample dosage, as explained in detail in Section 2.2.5, the CDW completely replaces the NA and the presence of ZCT has been discarded; however, the hydraulic reaction of the PC/CDW interface has been provided with the lowest mechanical strengths, as shown in Figure 4. If the average values of mechanical strength of the PCS and PC/ZCT specimens are compared with the PC/CDW, a difference in phase of 24.53 and 20.0 MPa, respectively, for each case.

Figure 5 shows the results of the calculation of the resistant activity index (RAI) applied to the normal values of mechanical strength to compression of the specimens analysed in the specific period of 28 days of curing. The percentage of minimum resistance required by Standard UNE-EN 196-1:2018 [32], known as RAI, corresponds to a value greater than or equal to 75% of the mechanical strength value of the referenced specimen (PCS), at 28 days of age.

According to Figure 4, the mechanical strength value of the PCS specimen at 28 days is 55.0 MPa, which represents the reference value by which the RAI of the remaining specimens were calculated. In the graph in Figure 5, there is a continuous green line which indicates the minimum percentage which is at 75%; all samples located above the green line comply with the RAI, according to the limits imposed by Standard UNE-EN 196-1:2018 [32], the opposite is true when the sample is below the line, as is the case in the PC/CDW specimen. Therefore, all the specimens analysed have a positive RAI except the PC/CDW, which makes these specimens ideal to carry out the objectives of this work.



**Figure 5.** Variations in the Resistant Activity Index (RAI) of specimens made with PC/ZCT, PC/CDW-ZCT (01 to 04) and PC/CDW, in relation to 75% of the value of the compressive strength of the PCS specimen.

#### 4. Conclusions

The chemical, physical and mineralogical analyses of CDW confirms that this material is ideal for recycling and reuse. However, the mechanical strengths are low when mixed only with the PC, in which case it does not comply with the RAI; nevertheless, this improves drastically when 25% of ZCT is incorporated into the mortar mixture. Based on the above, all proportions of the designed mixtures could be used, except in those made exclusively with PC/CDW.

Zeolitised cinerite tuff (ZCT) has a visibly complex mineral and chemical composition, however, its content in  $\text{SiO}_2$  and  $\text{Al}_2\text{O}_3$  as well as its cation exchange capacity (CEC), extremely large specific surface, and its abnormally reactive pozzolanic nature, among other properties, positively affect the hydraulic reactivity of the CDW and the gain of mechanical strength to compression over time.

The results of the mechanical strength tests have corroborated that construction and demolition waste (CDW) can favourably replace natural aggregate (NA), depending on the formulation of the dosage. The values of compressive strengths are within the parameters specified in the standard for the initial ages (7 days); however, they meet and even exceed in some cases the normal minimum mechanical strength at 28 days of age.

For production processes, two basic mixture proportions can be selected: which are the PC/CDW-ZCT-01 and PC/CDW-ZCT-02 specimens; however, since other specimens exceed the value of the RAI, mixtures of PC/CDW-ZCT-03 and PC/CDW-ZCT-04, could also be used. The use of these proportions of mixtures would represent a significant energy saving in the production process, a saving of 25% of PC and between 75 and 100% of NA.

Last, but not least, the mixing of CDW, NA, ZCT and PC could be a good solution to properly manage and use CDWs in urban environments, as well as to preserve industrial rock and mineral deposits.

**Author Contributions:** Conceptualisation, D.A.M., J.L.C., J.L.C.J. and L.P.; methodology, D.A.M., J.L.C., J.L.C.J. and L.P.; software, D.A.M., J.L.C., J.L.C.J. and L.P.; validation, D.A.M., J.L.C., J.L.C.J. and L.P.; formal analysis, D.A.M., J.L.C., J.L.C.J. and L.P.; investigation, D.A.M., J.L.C., J.L.C.J. and L.P.; resources, D.A.M., J.L.C., J.L.C.J. and L.P.; data curation, D.A.M., J.L.C., J.L.C.J. and L.P.; writing original draft preparation, D.A.M. and J.L.C.; writing review and editing, D.A.M., J.L.C., J.L.C.J. and L.P.; visualisation, D.A.M., J.L.C., J.L.C.J. and L.P.; supervision, J.L.C. and D.A.M.; project administration, D.A.M. and J.L.C.; funding acquisition, D.A.M. and J.L.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:** Not applicable.

**Acknowledgments:** The authors wish to thank the Laboratorio Oficial para Ensayos de Materiales de Construcción (LOEMCO) for the preparation of the samples, the performance of the tests and the interpretation of the results, as well as the financial support offered to cover the costs of translation of this work. The authors would also like to acknowledge the Laboratorio Centralizado of the Escuela Técnica Superior de Ingenieros de Minas y Energía (Universidad Politécnica de Madrid) for the SEM, XRF and XRD analysis. The authors send a message of thanks to Empresa de Proyectos Vértice, Departamento de Estructuras, in the city of Holguín (Cuba) for the technical support provided in this research.

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

## References

- Pacheco-Torgal, F.; Ding, Y.; Colangelo, F.; Tuladhar, R.; Koutamanis, A. *Advances in Construction and Demolition Waste Recycling: Management, Processing and Environmental Assessment*; Woodhead Publishing Series in Civil and Structural Engineering; Pacheco-Torgal, F., Ding, Y., Colangelo, F., Tuladhar, R., Koutamanis, A., Eds.; Woodhead Publishing: Sawston, UK, 2020; 592p, ISBN 9780128190555.
- Jesus, S.; Maia, C.; Brazão, C.; de Brito, J.; Veiga, R. Rendering mortars with incorporation of very fine aggregates from construction and demolition waste. *Constr. Build. Mater.* **2019**, *229*, 116844. [[CrossRef](#)]
- Azevedo, A.R.G.; Cecchin, D.; Carmo, D.F.; Silva, F.C.; Campos, C.M.O.; Shtrucka, T.G.; Marvila, M.T.; Monteiro, S.N. Analysis of the compactness and properties of the hardened state of mortars with recycling of construction and demolition waste (CDW). *J. Mater. Res. Technol.* **2020**, *9*, 5942–5952. [[CrossRef](#)]
- Silva, J.; de Brito, J.; Veiga, R. Recycled red-clay ceramic construction and demolition waste for mortars production. *J. Mater. Civ. Eng.* **2010**, *22*, 236–244. [[CrossRef](#)]
- De Oliveira, J.J.; Possan, E.; Zamboni, J.; Possebon, T.L. Evaluation of mechanical properties and carbonation of mortars produced with construction and demolition waste. *Constr. Build. Mater.* **2018**, *161*, 70–83. [[CrossRef](#)]
- Infante, R.; Brazão, C.; Rosário, V.; De Brito, J.; Faría, P.; Bastos, D. CO<sub>2</sub> sequestration by construction and demolition waste aggregates and effect on mortars and concrete performance—An overview. *Renew. Sust. Energ. Rev.* **2021**, *152*, 111668. [[CrossRef](#)]
- Fernández, E.; Jiménez, J.R.; Ayuso, J.; Fernández, J.M.; De Brito, J. Maximum feasible use of recycled sand from construction and demolition waste for eco-mortar production—Part-I: Ceramic masonry waste. *J. Clean. Prod.* **2015**, *87*, 692–706. [[CrossRef](#)]
- Saiz, P.; González, M.; Fernández, F.; Rodríguez, A. Comparative study of three types of fine recycled aggregates from construction and demolition waste (CDW), and their use in masonry mortar fabrication. *J. Clean. Prod.* **2016**, *118*, 162–169. [[CrossRef](#)]
- Katz, A.; Kulisch, D. Performance of mortars containing recycled fine aggregate from construction and demolition waste. *Mater. Struct.* **2017**, *50*, 199. [[CrossRef](#)]
- Duan, Z.; Hou, S.; Xiao, J.; Li, B. Study on the essential properties of recycled powders from construction and demolition waste. *J. Clean. Prod.* **2020**, *253*, 119865. [[CrossRef](#)]
- Duan, Z.; Hou, S.; Xiao, J.; Singh, A. Rheological properties of mortar containing recycled powders from construction and demolition wastes. *Constr. Build. Mater.* **2020**, *237*, 117622. [[CrossRef](#)]
- Jesus, S.; Pederneiras, C.M.; Brazão Farinha, C.; de Brito, J.; Veiga, R. Reduction of the cement content by incorporation of fine recycled aggregates from construction and demolition waste in rendering mortars. *Infrastructures* **2021**, *6*, 11. [[CrossRef](#)]
- Colangelo, F.; Cioffi, R. Mechanical properties and durability of mortar containing fine fraction of demolition wastes produced by selective demolition in South Italy. *Compos. B Eng.* **2017**, *115*, 43–50. [[CrossRef](#)]
- Ferreira, R.L.S.; Anjos, M.A.S.; Andreza, K.C.; Pereira, J.E.S.; Ledesma, E.F. The role of powder content of the recycled aggregates of CDW in the behaviour of rendering mortars. *Constr. Build. Mater.* **2019**, *208*, 601–612. [[CrossRef](#)]
- De Rossi, A.; Ribeiro, M.J.; Labrincha, J.A.; Novais, R.M.; Hotza, D.; Moreira, R.F.P.M. Effect of the particle size range of construction and demolition waste on the fresh and hardened-state properties of fly ash-based geopolymers mortars with total replacement of sand. *Process Saf. Environ. Prot.* **2019**, *129*, 130–137. [[CrossRef](#)]
- Sales, A.; Rodrigues, F. Concretes and mortars recycled with water treatment sludge and construction and demolition rubble. *Constr. Build. Mater.* **2009**, *23*–*26*, 2362–2370. [[CrossRef](#)]
- Del Río, M.; Vidales, A.; Piña, C.; Vitiello, V.; Santa Cruz, J.; Castelluccio, R. A review of the research about gypsum mortars with waste aggregates. *J. Build. Eng.* **2022**, *45*, 103338. [[CrossRef](#)]
- Antunes, M.L.P.; Botignon, A.; Oliveira, P.S.; Rangel, E.C. Utilization of gypsum from construction and demolition waste in Portland cement mortar. *Cerâmica* **2019**, *65* (Suppl. S1), 1–6. [[CrossRef](#)]
- Robayo-Salazar, R.A.; Valencia-Saavedra, W.; Mejía de Gutiérrez, R. 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](#)]
- Wu, H.; Zuo, J.; Zillante, G.; Wang, J.; Yuan, H. Status quo and future directions of construction and demolition waste research: A critical review. *J. Clean. Prod.* **2019**, *240*, 118163. [[CrossRef](#)]

21. Stefanidou, M.; Anastasiou, E.; Georgiadis, K. Recycled sand in lime-based mortars. *Waste Manag.* **2014**, *34*, 2595–2602. [[CrossRef](#)] [[PubMed](#)]
22. Asensio, E.; Medina, C.; Frías, M.; Sánchez, M.I. Fired clay-based construction and demolition waste as pozzolanic addition in cements. Design of new eco-efficient cements. *J. Clean. Prod.* **2020**, *265*, 121610. [[CrossRef](#)]
23. Shahmansouri, A.A.; Akbarzadeh Bengar, H.; Ebrahimi, E. Predicting compressive strength and electrical resistivity of eco-friendly concrete containing natural zeolite via GEP algorithm. *Constr. Build. Mater.* **2019**, *229*, 116883. [[CrossRef](#)]
24. Shahmansouri, A.A.; Nematzadeh, M.; Behnood, A. Mechanical properties of GGBFS-based geopolymers concrete incorporating natural zeolite and silica fume with an optimum design using response surface method. *J. Build. Eng.* **2021**, *36*, 102138. [[CrossRef](#)]
25. Standard UNE-EN 197-1:2011; Parte 1. Composición, especificaciones y criterios de conformidad de los cementos comunes. AENOR: Madrid, Spain, 2011.
26. Standard UNE-EN 933-1:2012; Ensayos para determinar las propiedades geométricas de los áridos. Parte 1: Determinación de la granulometría de las partículas. Método del tamizado. AENOR: Madrid, Spain, 2012.
27. Standard UNE-EN 933-9:2010; Ensayos para determinar las propiedades geométricas de los áridos. Parte 9: Evaluación de los finos. Ensayo de azul de metileno. AENOR: Madrid, Spain, 2010.
28. Standard UNE-EN 933-8:2012; Ensayos para determinar las propiedades geométricas de los áridos. Parte 8: Evaluación de los finos. Ensayo del equivalente de arena. AENOR: Madrid, Spain, 2012.
29. Standard UNE-EN 1097-6:2000; Ensayos para determinar las propiedades mecánicas y físicas de los áridos. Parte 6: Determinación de la densidad de partículas y la absorción de agua. AENOR: Madrid, Spain, 2000.
30. Standard UNE-EN 1744-1:2010; Ensayos para determinar las propiedades químicas de los áridos. Parte 1: Análisis químico. AENOR: Madrid, Spain, 2010.
31. Standard UNE 83-115-89; Áridos para hormigones: Medida del coeficiente de friabilidad de las arenas. AENOR: Madrid, Spain, 1989.
32. Standard UNE-EN 196-1:2018; Métodos de ensayo de cementos. Parte 1: Determinación de resistencias. AENOR: Madrid, Spain, 2018.
33. Tobón, J.I.; Payá, J.J.; Borrachero, M.V.; Restrepo, O.J. Mineralogical evolution of Portland cement blended with silica nanoparticles and its effect on mechanical strength. *Constr. Build. Mater.* **2012**, *36*, 736–742. [[CrossRef](#)]
34. Costafreda, J.L. Geología, Caracterización y Aplicaciones de las Rocas Zeolíticas del Complejo Volcánico de Cabo de Gata (Almería). Ph.D. Thesis, Universidad Politécnica de Madrid, Madrid, Spain, 2008.
35. Presa, L.; Costafreda, J.L.; Martín, D.A.; Díaz, I. Natural Mordenite from Spain as pozzolana. *Molecules* **2020**, *25*, 1220. [[CrossRef](#)] [[PubMed](#)]
36. Dai, X.; Li, X.; Wang, X. Morphology controlled porous poly(lactic acid)/zeolitic imidazolate framework-8 fibrous membranes with superior PM2.5 capture capacity. *Chem. Eng. J.* **2018**, *338*, 82–91. [[CrossRef](#)]
37. Korkuna, O.; Leboda, R.; Skubiszewska-Zie, J.; Vrublev's'ka, T.; Gun'ko, V.M.; Ryczkowski, J. Structural and physicochemical properties of natural zeolites: Clinoptilolite and mordenite. *Microporous Mesoporous Mater.* **2006**, *87*, 243–254. [[CrossRef](#)]
38. Nilchi, A.; Maalek, B.; Khanchi, A.; Ghanadi, M.; Bagheri, A.; Savoji, K. Ion exchangers in radioactive waste management: Natural Iranian zeolites. *Appl. Radiat. Isot.* **2006**, *64*, 138–143. [[CrossRef](#)]
39. Flanigen, E.M. Chapter 2 Zeolites and molecular sieves: An historical perspective. In *Introduction to Zeolite Science and Practice*; van Bekkum, H., Flanigen, E.M., Jacobs, P.A., Jansen, J.C., Eds.; Elsevier: Amsterdam, The Netherlands, 2001; Volume 137, pp. 11–35. [[CrossRef](#)]
40. Rodrigues, F.; Carvalho, M.T.; Evangelista, L.; De Brito, J. Physical–chemical and mineralogical characterization of fine aggregates from construction and demolition waste recycling plants. *J. Clean. Prod.* **2013**, *52*, 438–445. [[CrossRef](#)]
41. Rosell-Lam, M.; Villar-Cocíña, E.; Frías, M. Study on the pozzolanic properties of a natural Cuban zeolitic rock by conductimetric method: Kinetic parameters. *Constr. Build. Mater.* **2011**, *25*, 644–650. [[CrossRef](#)]
42. Silva, R.V.; De Brito, J.; Dhir, R.K. Properties and composition of recycled aggregates from construction and demolition waste suitable for concrete production. *Constr. Build. Mater.* **2014**, *65*, 201–217. [[CrossRef](#)]
43. Da Silva, S.R.; De Oliveira, J.J. Investigation of mechanical properties and carbonation of concretes with construction and demolition waste and fly ash. *Constr. Build. Mater.* **2017**, *153*, 704–715. [[CrossRef](#)]