



# **Potential Use of Oyster Shell Waste in the Composition of Construction Composites: A Review**

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Abstract: The oyster shell is a residue rich in calcium carbonate, which can be reused as a raw material for creating building materials. For this reason, many researchers focused on the incorporation of oyster shell in the composition of composites, as it is a means of contributing to the economic sustainability by reducing the presence of pollution caused by aquaculture waste in the environment, thus increasing the value chain of the construction sector and reducing its carbon footprint. This paper intends to systematize the scientific production related to oyster shell-based composites in construction, carrying out a search using the Scopus tool and a systematic review based on the PRISMA statement. The results show that research on the incorporation of oyster shell into cementitious mortar mixtures, with a focus on its use in concrete, dominates existing scientific research. There is a lack of studies on the incorporation of the oyster shell that address its application as an aggregate or binder in the composition of coating and laying mortars. Most existing research is from Asia, and there is a lack of research in some parts of Europe. In the Americas, Africa and Oceania, no existing studies were found. Despite the growing understanding of the importance of sustainability and economic issues related to products used in the blue circular economy sector, there are still few studies that consider the incorporation of waste or by-products of aquaculture. Future investigations that cover these practical and contextual gaps can contribute to the better use of oyster shell waste and its insertion in the blue circular economy.

Keywords: sustainability; mortar; oyster shell; performance; review; blue circular economy

## 1. Introduction

According to the report "The State of World Fisheries and Aquaculture" (SOFIA), which was published by Food and Agriculture Organization of the United Nations (FAO) in 2020, fish produced via aquaculture represented 87.5 million tons of annual seafood production. The worldwide production of bivalve molluscs represented 20% of the total produced considering inland and marine and coastal aquaculture, which is equivalent to 17.7 million tons [1].

Oysters are bivalve organisms composed of more than 95% of calcium carbonate, and 5 million tons of related waste are illegally disposed every year [2,3]. Oyster Shell Waste (OSW) causes serious problems in the oceans due to its disposal in large quantities [4], as it silts and eutrophicates in bays [3], and even creates odors, undergoes microbial decomposition, and attracts insects when disposed via landfill [5,6]. These situations cause environmental pollution and limit of quality of life for populations close to these places [7].



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**Copyright:** © 2023 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/). Material recycling plays a crucial role in overcoming current environmental problems. It involves the process of transforming waste into new products, thereby reducing the need for natural resources and decreasing both the amount of waste disposed in landfills and pollution [8–17].

The negative impacts of incorrectly disposing of oyster shells can, thus, be minimized via their reuse. The construction sector is an important industry for the reuse and recovery of resources from other industries. In China, many historic masonry buildings in the coastal areas of Fujian province have coatings made with oyster shell clay mortar, and it is advisable, if possible, to use the same material to repair mortars. In Portugal's Algarve region, as can be seen in photographs from the 1950s, fishermen traditionally incorporated this type of waste into the construction of their houses [18]. For those reasons, there is also a sociocultural aspect that can be valued and re-integrated into the productive sector of coastal regions, generating wealth, as well as obvious environmental benefits, such as a reduction in both the carbon footprint generated by construction and waste. In addition, the Portuguese Government's recommendations emphasize the use of local materials for the delimitation of aquaculture cultivation ponds. Despite these sanitary requirements, the amount of waste produced during explorations is often not sent in its entirety for incineration, which generates environmental impacts. For those reasons, it is necessary to create solutions for its valorization.

Based on the UN Sustainable Development Goals (SDGs) 2030, fisheries and aquaculture are fundamental alternatives within the scope of sustainable development of oceans and seas (SDG 14). According to FAO data [1], fishery and aquaculture waste is estimated to reach 106 million tons in 2030 (an overall growth of 22%, or nearly 19 million tons, compared to 2020). Shell waste is used in different contexts in the civil construction field. Therefore, developing sustainable practices for this sector is essential to face up to the social, economic and environmental problems to which the SDGs refer [1].

Previous research with literature reviews focused on studies of the use of shell residues in concrete [7,19–21]. For example, Ruslan et al. [21] analyzed the use of OSW as a substitute for cement and, partially, for sand in the composition of concrete. The cited authors addressed questions about processing methods and the fresh and hardened properties of concrete produced with OSW.

Mo et al. [7], in turn, carried out a study of previous research on the use of shell residues as partial substitutes for conventional materials in concrete and other related cement-based products. The characteristics of different types of shell residues were discussed, as well as their incorporation's effects on the fresh and hardened properties of concrete.

Eziefula et al. [20] carried out a literature review study on concrete with partial and total replacement of fine and coarse aggregates by seashells. The article was divided into the stages covering the physical, mechanical, and chemical properties of the shells, followed by a discussion of the physical and mechanical properties, durability, sound absorption, and thermal insulation of concrete (in both fresh and hardened states). The authors also commented on possible applications of seashells in the construction industry.

Prusty et al. [19] also carried out a study of the literature discussing some agro-waste materials that are used as partial substitutes for fine aggregate in concrete, self-compacting concrete, and mortar. The initial topics reported included the physical and chemical properties of these residues. The proportion of concrete mixtures and fresh and hardened properties were also discussed. Durability, thermal conductivity, and cyclic behavior were also addressed.

Zhan et al. [22] provided a review of the reuse of shell waste to promote sustainable shellfish aquaculture. This review summarizes research on the reuse of shell waste, including applications in the areas of agriculture, construction, environmental protection, chemical industry, food additives, and biomaterials.

The research progress on the applications of seashell adsorption behaviors in cementbased materials was studied by Li et al. [23]. The authors reviewed the principles of adsorption (kinetics and isotherms) and discussed the effects of pH, contact time, temperature, pollutant concentration, and other factors on the adsorption of heavy metal ions and basic dyes to shells. Finally, the relevant applications of shells in the field of construction (cement mortar, concrete, and architectural coatings) are reviewed.

Large-scale research was carried out regarding the reuse of discarded shells to replace conventional materials [7]. In the civil construction industry, studies of the incorporation of oyster shell wastes can be found for concrete [24–27], brick [28,29], hot-mix asphalt [30], artificial stone [31], soil mixing and drainage layers [32,33], magnesium phosphate cement [34], and residential finish [35]. This range of research demonstrates the great potential of using shell residues in different contexts in the field of civil construction, which motivates the present study. This paper updates the studies carried out regarding the incorporation of oyster shell residue in the composition of construction composites, identifying other lines of research for OSW with different applications within construction composites (e.g., mortars). Moreover, the preparation of the material, its behavior when incorporated into composites, and the main insights offered by the relevant literature are systemized and discussed.

## 2. Research Methodology

The objective of the research was to map the production of scientific studies on the potential use of OSW residue in mortars. For this study, a database was selected—Scopus and the same filters were applied to all searches. In the document type filter, the following options were selected: article and book chapter; in source type, journal, book series, and book were selected; and English was the only language selected. Through the Reporting Items for Systematic reviews and Meta-Analyses (PRISMA) statement [36], the systematic literature review was based on a search for common themes among the scientific articles found, as well as the framing based on these themes [37], thus mapping the main lines of research for scientific production in the area.

#### 3. Description of the Study and Results

The keywords were chosen according to the subject of study. Word combinations resulted in the search of 17 different groups of oyster material usage. The identification of each group, along with the results, is shown in Table 1.

Group	Keywords	Number of Documents
1	Oyster shell; construction; waste	18
2	Mortar; calcination; shell	7
3	Mortar; composition; shell	46
4	Mortar; shell; oyster	34
5	Mortar; oyster; properties	20
6	Mortar; oyster; strength	19
7	Mortar; oyster; durability	6
8	Mortar; oyster; sustainability	3
9	Mortar; oyster; environment	4
10	Mortar; oyster; waste	16
11	Mortar; oyster; recycling	2
12	Mortar; lime; shell	36
13	Mortar; mollusc; sustainability	2
14	Mortar; lime; oyster	7
15	Mortar; cement; oyster	17
16	Oyster; shell; sand; mortar	11
17	Mortar; oyster; powder	11
Total		259

 Table 1. Scopus search engine results.

All studies published up to the first half of 2022 were considered. A total of 259 studies were found by the different research groups. After the rejection of repeated articles, the number of studies was reduced to 111. After this first screening, articles in the area

of chemistry were found that point out relevant questions regarding the treatment and composition of the oyster shell, encouraging its use in construction applications [38]. Articles with residues from the food sector [39], as well as other aquaculture residues [40–43] used in the development of building materials, were also identified. However, as they did not correspond to the use of the type of residue that was the focus of this study (oyster shell), they were excluded, leaving 33 remaining studies. Studies of the incorporation of oyster shell residues in other construction materials, such as concrete (four studies), brick (two studies), hot-mix asphalt (one study), artificial stone (one study), soil mixing and drainage layer (two studies), magnesium phosphate cement (one study), and residential finish (one study), were also excluded, because the focus of the study was analyzing the potential use of oyster shell residue in mortars. Although these studies are not addressed, they show that there is increasing interest in using oyster shells in the production of different construction materials.

The next step was to identify only the use of OSW in composites (coating, laying and concrete mortars, and paste), and 21 studies remained; therefore, 19 studies were used in this research.

The research methods included the following type of analyses: quantitative, lines of research, and descriptive. Quantitative analysis contributed to the following knowledge topics: the number of publications over time, type of publication journals, location, and geographic distribution of publications. Power Business Intelligence (Power BI, 2023) software was used as a support tool to demonstrate geographic distribution by region. To assist in the analysis of the lines of research, the Word Clouds tool was used. In the case of the descriptive analysis, different subjects found in the studies were addressed. The minimum number considered for occurrences of a theme was five, resulting in the descriptions of 12 different subjects. The descriptions of the studies are given in Section 3.3.

#### 3.1. Quantitative Analysis

## 3.1.1. Number of Publications over Time

The research carried out in the Scopus database did not include a temporal threshold. The oldest publication found on the use of oyster shell in composites was published in 2003. The first studies found developed with the use of oyster shell with sand replacement sand. After 2012, research began to use oyster shell as a powder to replace the binder, and they are the studies with the highest number of published papers.

The amount of papers published since 2020 using OSW increased by 78% compared to the time period ranging from 2000 to 2010. The years that presented a higher number of publications were 2019 and 2021, with a total of four studies, making up around 21% of the total. The number of publications increased each year, with the exception of the year 2020, which had one less publication than the previous year. The distribution of papers as a function of time can be seen in Figure 1.

The data indicate a growing interest in studies on the use of oyster shells in the composition of mortars, especially after September 2015, when the United Nations launched the 2030 Agenda. This relationship between the achievement of UN goals and the increase in interest in research on the use of oyster shells in civil construction may be the subject of future research.

Moreover, the potential development of bivalves in the aquaculture subsectors remains significant, especially in the marine environment, having acquired a positive perception among consumers as a healthy and sustainable food option. In 2020, global exports of bivalve molluscs totaled US\$4.3 billion, representing about 2.8% of the value of global exports of aquatic products. Aquaculture of bivalve molluscs is certainly important in the Americas, Europe, Asia, and Oceania [1].

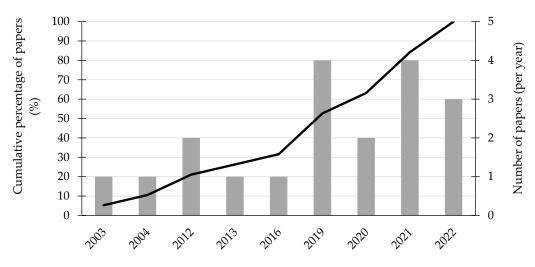


Figure 1. Distribution of publications as a function of time.

## 3.1.2. Type of Published Journals

The 19 studies found were published in 16 different journals (Table 2). The periodicals cover the areas of construction and materials, sustainability, and chemistry. The journals comprise the following periodicals: the Journal of Cleaner Production, Case Studies in Construction Materials, Materials, and Construction and Building Materials; each journal had two publications (10.5%). The rest of the journals each had one publication (5%).

Table 2. Types of publishing journals.

Journal Title	Number of Studies	Areas of Expertise
Sustainability (Switzerland)	1	Sustainability
Advanced Powder Technology	1	Chemical engineering
Journal of Cleaner Production	2	Environmental sciences
Waste Management and Research	1	Management of and research into residues
Waste Management	1	Environmental sciences
Case Studies in Construction Materials	2	Civil engineering
New Journal of Chemistry	1	Chemistry
Materials	2	Science and materials engineering
Buildings	1	Construction science, building engineering, and architecture
AMB Express	1	Applied and industrial microbiology
Asian Journal of Civil Engineering	1	Civil engineering
Materiales de Construccion	1	Construction, science, and materials technology
Journal of Environmental Management	1	Environmental sciences
Jiegou Huaxue	1	Chemistry
Construction and Building Materials	2	Civil engineering

3.1.3. Location and Geographic Distribution of Publications

The investigation carried out did not eliminate any country. The research revealed that much of the scientific production on the subject is concentrated in Asia (89%). China leads in terms of the number of published surveys, making up a total of 47% of the studies, followed by South Korea with 21% of the studies, Taiwan with 16% of the studies, and Thailand with 5% of the studies. A total of 11% of the observed studies are from Europe. European surveys are concentrated in France. Figure 2 depicts a heat map produced using Power BI.



Figure 2. Geographical distribution of publications about use of oyster in composites.

The largest number of studies being produced in China is also due to the fact that it has been the largest producer of aquatic animals since 1991. In 2020, it was the global market leader, representing 56.7% of the quantity of aquatic animals produced worldwide (similar trends were observed in recent years). However, in some large oyster-producing countries, in terms of the cultivation of all marine species, bivalves account for a high percentage of the total aquaculture production. These countries include New Zealand (86.9%), France (75.4%), Spain (74.8%), South Korea (69.7%), Italy (61.6%), and Japan (51.8%); the world average figure is 18.4%.

With the exception of Africa, which registered a decline of 1.2%, all other regions experienced continued growth in the aquaculture sector in 2020. Chile, China, and Norway were the top producers in the Americas, Asia, and Europe, respectively [1].

According to the FAO [1], the most important species of bivalve molluscs for international trade are scallops, clams, oysters, and mussels, while the vast majority of bivalve molluscs consumed are cultivated and produced in several European countries, North America, China, and Chile.

However, the number of countries that incorporate oyster shell waste into research is still small. Given the growing understanding of the sustainable and economic issues related to products ued in the blue circular economy, this aquaculture waste has great potential for application in the construction sector.

## 3.2. Analysis of Research Lines

## Incidence of Keywords

Using the Word Clouds tool, Figure 3 represents the incidence of keywords of all analyzed articles. The word "Mortar" was the one that appeared the most, being present in nine articles as a keyword.

The term "durability" appears as a keyword in four articles. As predicted, several words that were also frequently repeated were "waste oyster shell" and "strength", appearing a total of three times each. Other terms, such as "supplementary cementitious materials", "properties", "mechanical property", and "compressive strength", appeared in two articles. The other words were cited once in the studies.

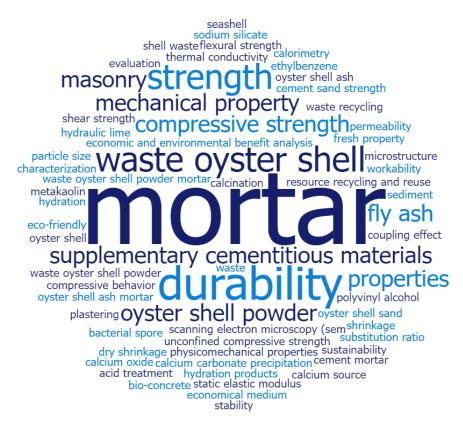


Figure 3. Incidence of keywords.

The incidence of keywords helps us to understand which subjects are most often debated in articles referring to the use of oyster shell in the composition of mortars (for structural and non-structural applications).

## 3.3. Descriptive Analysis

## 3.3.1. Research Lines

The descriptive analysis searched for common themes found among the scientific articles, and these themes were grouped within lines of research (treatment and application of oyster shell in mortar composition; properties in the fresh state; properties in the hardened state; eco-efficiency).

Considering 100% of the articles (19 studies), the most prominent theme was mechanical strength, which was addressed in 95% of the studies. The residues treatment process before use, such as washing (79% of studies), drying (68% of studies), and grinding to the necessary particle size (as binder or aggregate) for application in mortar composition, was the second most discussed item, being the subject of 84% of the studies. The properties in the fresh and hardened states of the produced mortars were the next most discussed topic in the studies. Workability and microstructure were discussed in 42% of the studies. Heat of hydration/setting time, volume of permeable void, and diffusion coefficient of chloride ions were the next most discussed subjects (32% of the studies). The other properties in the hardened state, such as the volume of permeable void, water absorption, shrinkage, and water permeability, were present in 26% of the studies. The topic of eco-efficiency was also addressed in 26% of the studies. The total number of articles that commented on each theme can be seen in Table 3.

The results show that mechanical resistance was the topic of greatest interest in the articles that proposed studying the incorporation of oyster shell into the composition of mortars, as well as how this residue affects mortar behavior. These details are discussed in Section 3.3.8.

Search Line	Subject	Number of Articles
Treatment and application of the	Washing	15
oyster shell in mortar	Drying	13
composition	Grinding	16
Proportion in the fresh state	Workability	8
Properties in the fresh state	Heat of hydration/setting time	6
	Mechanical	18
	Shrinkage	5
	Water absorption	5
Properties in the hardened state	Volume of permeable void	5
	Permeability coefficient	5
	Microstructure	8
	Diffusion coefficient of chloride ions	6
Eco-efficiency		5

Table 3. Search line, items, and numbers of studies.

3.3.2. Treatment and Application of the Oyster Shell in the Mortar Composition

Prior to the practical use of OSW, the oyster shell was subjected to some initial treatment. In the analyzed studies, it was observed that 100% of the studies indicated some important treatment steps before reusing shell residues, such as washing, drying, calcining, and grinding to the desired size [7].

These types of treatment refer to when oyster shell is incorporated, as a powder or aggregate, in the composition of composites. Figure 4 shows an example of grinding oyster shell to create aggregate and powder.



Figure 4. Grinding oyster shell to create aggregate (0-4 mm) and powder (150 µm).

#### 3.3.3. Washing and Drying

The washing process was performed with water in 74% of the studies. Some studies did not report and/or did not mention whether the residue was washed (21%). Nonetheless, one study carried out the washing of the material after grinding [44], while another study used the oyster shell soiled with clay and sand [45]. Seo et al. [46] reported leaving the shell submerged in water for approximately one week to remove salts and other impurities. Weng et al. [47] discussed boiling the shells until the pH and conductivity of the boiled water was approximately equal to that of pure water.

Washing becomes an important step in the process of preparing oyster shell waste to be used in research. This step is essential because, in this process, superficial and foreign substances, such as salt content and traces of organic matter, are removed [44,46,48,49]. Authors such as Ez-zaki et al. [48] measured the amount of free chloride in the final products, and verified that the washing process used significantly reduced the chloride content in the residue.

Based on the results of the study by Ez-zaki et al. [50], the waste pre-treatment process showed significant results. The washing step decreased the amount of residual chloride trapped in the waste, and the thermal process led to the transformation of clay minerals. For the material drying, this step took place immediately after washing. One study mentioned that this process was carried out naturally [46]. However, in total, 58% of the studies carried out this procedure using ovens to store and dry the residue. The temperatures and drying times ranged from 45 to 150 °C and 2 to 72 h, respectively. Chen et al. [44] report that at  $110 \pm 5$  °C for 24 h, it is possible to remove water and organic matter from the shells. A total of 32% of the studies used this time range. In 37% of the studies, no information was reported about this process. There is also one study in which drying was the last step, performed only after the calcination process, grinding, and quicklime production [45].

## 3.3.4. Grinding and Use of Oyster Shell as Powder

In 58% of the studies, total or partial replacement of the binder by OSW powder was carried out. The oyster shell was collected in its natural format, and the main chemical composition of the oyster shell was calcium carbonate (CaCO<sub>3</sub>), reaching the amount of 96% by weight [49]. When oyster shell residue powder is used to replace the binder, the residue, the main component of which is CaCO<sub>3</sub>, can be heated to high temperatures (greater than 800 °C) and transformed into lime (CaO). This process is known as calcination [51], which was performed in 64% of the studies that replaced the binder with OSW powder. In cement mortar, the percentage substitution of binder by OSW powder was between 3 and 33%. Liao et al. [51] stated that it is reckless to incorporate calcined OSW powder as the only binder in mortar due to the negative effect on mechanical strength and other properties for concrete. As a partial substitute for cement, powdered oyster shell residue was used in 64% of the studies that aimed to replace the binder.

However, in the case of using mortar for laying and walls coating (36% of the studies), these mortars had 100% of the binder as powder from the residue of oyster shells. In half of these papers, information was given explaining that the material was calcined between 800 and 1200  $^{\circ}$ C, while in the other half of these papers, this information was not disclosed.

Table 4 presents the details of the preparation for using the OSW, information on the composition of the oyster shell, and the percentages of residue used in the composition of mortars when the binder was replaced.

Research	Shell Preparation Process	Composition of the Shell	Materials Used in the Mix	Quantity of Replacement	Objective
Bin-Yang et al. [52]	Washing, drying, and wet grinding, followe by drying at 105 °C (for 24 h) and sieving (165 µm)	CaO: 54.31%	Cement: oyster shell, powder, sand, and water	5, 10, 15, and 20%	Cement mortars for concrete
Lertwattanaruk et al. [53]	Washing, drying, coarse grinding (4.47 mm), wet grinding for 3 to 4 h (0.075 mm), and drying at $110 \pm 5$ °C (for 24 h)	CaO: 53.59%; CaCO <sub>3</sub> : 96.8%	Cement Type I: sand-ground seashell (short-necked clam shell, green mussel shell, oyster shell, and cockle shell)	5, 10, 15, and 20%, by weight	Cement mortars for concrete
Ez-zaki et al. [48]	Washing, decanting, drying at 40 °C, grinding (200 μm), and calcination between 650 and 850 °C for 5 h	CaO: 48%	Cement Type I 52.5: oyster shell, powder, sediments, sand, and water	8, 16 and 33%, by weight	Pastes and mortars of cement for concrete

Table 4. Details of residue used as a binder.

Research

Ez-zak et al.

[50]

Seo et al. [46]

Materials Used in the Mix	Quantity of Replacement	Objective
Cement Type I 52.5:oyster shell, powder, sediments, sand, and water	8 and 33%, by weight	Pastes and mortars of cement for concrete
Cement Type I: oyster shell, powder, sand, and water	3, 6, 9, and 12%	Cement mortars for concrete

## Table 4. Cont.

Composition of

the Shell

Predominantly

formed of calcium

carbonate CaCO<sub>3</sub>

CaO: 98% (after

calcination)

**Shell Preparation** 

Process

Washing, decanting, drying at 40 °C,

grinding (200 µm), and

calcination between 650

and 850 °C for 5 h Soaking in water (1 week), washing, air

drying, grinding,

calcination at 1000 °C

(for 3 h), and grinding (150 µm)

Zhang et al. [45]	No cleaning; grinding; calcination at 800, 1000, and 1200 °C for 2 h to produce quicklime; grinding (1 mm); soaking for 120 h to produce burnt lime; drying at 105 °C; and grinding to powder	CaCO <sub>3</sub> : 97.1% (before calcination)	Oyster shell, powder, sand, water, and glutinous rice	100%	Quicklime mortar for coating mortars
Chen et al. [54]	-	-	Oyster shell ash/sand/clay/water (new construction) and oyster shell ash/siliceous material A/aluminous material B/sand/water (masonry reinforcement)	100%	Laying mortars for new construction and masonry reinforcement
Weng et al. [47]	Washing, boiling, grinding (200 mesh), and calcination at 1050 °C for 90 min	CaO: variations from 56.04 to 75.23%	Oyster shell, powder, and water	100%	Lime paste for ancient cultural relics
Chen et al. [55]	-	-	Oyster shell ash/sand/clay/water (new construction) and oyster shell ash/siliceous material A/aluminous material B/sand/water	100%	Laying mortars for new construction and masonry reinforcement
Liao et al. [51]	Washing, drying at 100–110 °C (for 24 h), grinding (0.6 mm), calcination at 950 °C for 2 h, and grinding (10 to 60 µm)	CaCO <sub>3</sub> : 85.16%	Cement 42.5R: metakaolin–oyster shell, powder, sand, water, and superplasticizer	5, 8, and 10%	Cement– metakaolin mortar for composites
Liu et al. [56]	Washing, drying at $105 \pm 5$ °C (for 24 h), calcination at $850-950$ °C for 2 h, and grinding (0.5 to 40 $\mu$ m). The mesh sizes chosen for sieving were 1250, 3000, or 6000	CaO: 46.6%	Cement 42.5: oyster shell, powder, lithium, slag, ground granulated blast furnace slag, sand, and water	10%	Cement mortar for green concrete

Weng et al. [47] used OSW powder as the only binder. The particles were crushed through a 200 mesh sieve (0.075 mm), and the calcination process was then carried out. Zhang et al. [45], in addition to an initial grinding, which occurred after the calcination step, performed grinding and sieving again, with the particle size of the oyster shell reduced to less than 1 mm. Two other studies that also used OSW powder as a binder did not comment on particle size.

In other studies that carried out partial substitutions of the binder by OSW powder, namely the studies by Ez-zaki1 et al. [48] and Ez-zaki et al. [50], the particles were reduced to a size of 75  $\mu$ m, and after this process, they were calcinated. Lertwattanaruk et al. [53] performed a coarse grinding to 4.47 mm, and after a wet grinding with water in a ball mill for 3 to 4 h, they also reduced the oyster shell particles to 75  $\mu$ m (in this study, the authors did not mention the calcination stage). In the study of Bin-Yang et al. [52], the diameters of the particles were reduced until they passed through the mesh of 165  $\mu$ m (in this study, the residue was not calcined at any time). Liao et al. [51] initially ground the residue with a crusher until it passed through a sieve of 0.6 mm; after that, it was calcined, and the calcined residue then went through a ball mill treatment (the diameter of the balls was 40 mm, the number of balls was 37, and the volume of the mill was 5 kg) for about 1-2 h. Finally, the powder particles used were those that passed through the 60  $\mu$ m sieve. In the study of Liu et al. [56], after calcining the shells, the grinding process was carried out until the particles had dimensions between 0.5 and 40  $\mu$ m (the size of the particles varied being: 1250, 3000 or 6000 mesh). Seo et al. [46] reduced the particles to 150  $\mu$ m powder, performing the grinding and crushing again after calcination.

Lertwattanaruk et al. [53], when using a wet ball mill to grind sea shells into relatively fine particles comparable to Portland cement, obtained an average oyster shell size of 13.93  $\mu$ m, compared to 22.82  $\mu$ m for Portland cement. Bin-Yang et al. [52] described, in their conclusions, the type of oyster shell grinding that has an effect on the strength of cementitious mortar. The authors reported that wet ball milling produces a much finer, more evenly distributed oyster shell powder than that obtained via dry ball milling.

Other authors used oyster shell as an alternative source of nutrients in water (for mortar composites such as bioconcrete), though the granulometry powder used was not revealed [57].

#### 3.3.5. Grinding and Use of Oyster Shell as Aggregate

Regarding the replacement of the fine aggregate by OSW, seven studies were found. As an aggregate, the maximum granulometry sieve found in the studies (two studies) was 5 mm [44,58]. Liao et al. [59] used a jaw crusher and a ball mill to reduce the grain size until it was less than 0.6 mm. Yoon et al. [2] separated the ground residue into large particle size bands of 4.75–2 mm and small particle size bands of 2–0.074 mm. Wang et al. [60] reported the use of oyster shell as an aggregate in the mortar composition, and this sand met the specifications for fine aggregates (ASTM C33 and ASTM C136). Yoon et al. [5] analyzed four types of different particle sizes and used the particle size that had a D60 of 1.4 and a D30 of 0.95 mm. Furthermore, in one of the studies, the OSW was crushed to 0.6 mm and received treatment with polyvinyl alcohol (PVA) and sodium silicate (SS) to try to improve its performance and durability characteristics [49].

The details regarding the preparations and investigated compositions used when the residue replaced the aggregate are presented in Table 5.

Research	Shell Preparation Process	Composition of the Shell	Materials Used in the Mix	Quantity of Replacement	Objective
Yoon et al. [5]	Crushing and separation into four types of granulometry	CaCO <sub>3</sub> is approximately 96%	Cement: soil, sand, oyster shell, and water	20, 40, 60, and 80%	Cement mortars (resources made from pure calcareous materials)
Yoon et al. [2]	Washing, drying at 105 °C, crushing, and separation into large (4.75–2 mm), and small particles (2–0.074 mm)	Oyster shell is primarily composed of naturally formed calcium carbonate (CaCO <sub>3</sub> ). CaO = 52.94%	Cement: fly ash, ethylbenzene, sand, and oyster shell	100%	Cement mortars for concrete
Wang et al. [60]	Washing and grinding	CaCO <sub>3</sub> is 96% by weight	Cement Type I C150: sand, oyster shell, fly ash, water, and superplasticizer	5, 10, 20, and 30%	Cement mortars for concrete
Chen et al. [44]	Crushing (5 mm), washing, and drying at 110 °C (for 24 h)	CaCO <sub>3</sub> with high content	Cement Type I 42.5: fly ash, blast furnace slag, sand, oyster shell, water, and superplasticizer	30%	Cement mortars for concrete
Liu et al. [49]	Cleaning, drying at 100–110 °C (for 24 h), grinding (0.6 mm), and treatment with polyvinyl alcohol and sodium silicate.	-	Cement 42.5: sand, oyster shell, water, and superplasticizer	20%	Cement mortars for concrete
Liao et al. [58]	Washing, drying at 105 °C (for 5 h), and grinding (5 mm)	CaO = 60.16%	Cement Type I 42.5: sand, oyster shell, water, and superplasticizer	20%	Cement mortars for concrete
Liao et al. [59]	Washing, drying at 105 °C (for 5 h), crushing in jaw crusher and ball mill (0.6 mm), and drying at 105 °C for 8 h	CaCO <sub>3</sub> is more than 95%	Cement 42.5: metakaolin, sand, oyster shell, water, and superplasticizer	10%, 20%, and 30% by volume	Cement mortars for concrete

Table 5. Details of residue used as an aggregate.

## 3.3.6. Workability

According to Seo et al. [46] and Liao et al. [51], the hydration of calcinated OSW powder can induce the loss of workability by increasing the consistency of fresh mortar, which occurs due to the greater amount of water consumed for the formation of Ca(OH)<sub>2</sub>. Even so, Seo et al. [46] reported the same workability between the reference mortar and the mortars that had the cement replaced by the OSW powder calcined (3, 6 and 9%) at 3 min after mixing, except when replaced at 12% (the highest percentage chosen). Nonetheless, at 40 min, the workability presented was approximately 90% of the figure obtained at 3 min, indicating that the incorporation of calcined oyster shell powder did not result in a rapid loss of workability. In the case of the study by Liao et al. [51], the authors replaced 5, 8, and 10% of the cement with calcined oyster shell powder and found that the greater the amount of replacement, the lower the workability of the mortar. This study also used the replacement of 10% of the cement with metakaolin. However, the workability results decreased between 100 and 120 mm one hour after mixing, which is consistent with the established limits.

On the other hand, Lertwattanaruk et al. [53], when replacing the cement with oyster shell powder with particle sizes smaller than 0.075 mm (without calcination), noticed an increase in the free water content in the mixture due to the decrease in the amount of cementitious material present. The higher the replacement percentage (5, 10, 15, or 20%), the more visible this behavior becomes. According to the authors, as the replacement rate increased, there was a decrease in water retention due to the particles being flat and of low porosity. As a result, there was less friction between the residue and the cement particles, which generated a reduction in water demand and improved workability. This study used other marine residues and reported that oyster shell residue had the greatest need for water, due to its relatively high blain-specific surface area (oyster:  $14,280 \text{ cm}^2/g$ ; clam:  $8279 \text{ cm}^2/g$ ; green mussel:  $6189 \text{ cm}^2/g$ ; cockle shell:  $8299 \text{ cm}^2/g$ ). Thus, more water was needed to encase the oyster shell particles than was needed for the other types of shells.

On the other hand, when sand was replaced by OSW, a greater effect of friction between the residue and the cement particles was recorded due to the irregular surface of the residue, which reduced the fluidity of the mortars produced [60]. The authors also comment that a high internal porosity may also have influenced this outcome. With irregularly flat particles, the filling water content between the particles is deficient and mixing friction increases. In the case of internal pores, they can absorb moisture during the mixing process, thus increasing the water consumption of the mortar. Other authors also observed that as the rate of replacement of river sand by oyster shell residue increases, the cement mortar slump decreases [58,59]. Liao et al. [59] mentioned that the decrease in workability was attributed to the high rate of water absorption of the mortar. Wang et al. [60] found the absorption rate for oyster shell sand to be 7.66%, i.e., approximately 3.2 times that of the fine aggregate (2.38%), and that this issue can cause difficulties in compaction. The high rate of water absorption was also the reason for the loss of fluidity indicated in the study by Ez-zaki et al. [50], who replaced percentages by weight of cement with calcined OSW.

Liao et al. [58] replaced river sand with OSW in different particle size ranges: fine, medium, and coarse. In terms of workability, this parameter was significantly influenced by granulometry, and the results of the cement mortars with the residue with fine and medium granulometry were reduced by 17.14% and 9.28%, respectively, in relation to the reference mortar. The authors reported that workability performance was reduced due to the more irregular surface and higher specific surface area of the particles (coarse: 1284 kg/m<sup>3</sup>; medium: 1299 kg/m<sup>3</sup>; fine: 1354 kg/m<sup>3</sup>), as finer particles absorb more water and decrease the free water of the mixture. The coarse residue showed higher slump and lower slump loss rates. After verifying this finding, a mixture was produced using superplasticizer additive, and the workability was improved as a function of the use of this product. Nonetheless, both mixtures reached the requirements recommended by the standard.

## 3.3.7. Heat of Hydration/Setting Time

According to Liao et al. [58], when using coarse OSW particles as a substitute for the aggregate, the initial and final setting times were longer. This statement was proven through setting-time tests, where the values obtained were 516 min, 492 min, and 464 min for cement mortars produced with coarse, medium, and fine aggregates, respectively. Wang et al. [60], in addition to replacing sand with OSW, used fly ash in the mixtures and achieved prolonged setting times for the mortar. The most extensive time was found when 30% OSW was incorporated (initial setting time: 83 min; final setting time: 180 min). In this case, this result may occur due to the use of fly ash that releases low levels of calcium oxide, as well as the Ca(OH)<sub>2</sub> being consumed in the mortar during the hydration period. As a result, the reaction is reduced, and the setting times increased.

Lertwattanaruk et al. [53], when replacing cement with OSW powder, found a rise in initial setting times that was within the range indicated by the standards ( $\geq$ 60 min). As a result, mortars containing ground OSW had a longer setting time, which are an advantage for plastering in hot climates. When the OSW powder was calcined, the authors Seo et al. [46] found, during the hydration process, that the initial heat of hydration increased due to dissolution of CaO and formation of Ca(OH)<sub>2</sub>, slightly affecting the rate of formation of C–S–H at the initial stage of the reaction, and leading to a decrease in heat evolution at a later stage. Ez-zakia et al. [48], when replacing cement with calcined oyster shell powder and other sediments, achieved optimal replacements in the range of 8 to 16% with good hydraulic reactivity. In this case, the hydraulic reactivity of the additions due to the presence of metakaolin, gismondine, and the CaO, which were also due to the high fineness of the particle size of the treated sediment and the calcined OSW powder, caused a large calorimetric effect.

The authors Chen et al. [54] report that in China, the oyster shell ash mortars have some disadvantages, which occur due to the fact that they have a slow hardening time after application. From this perspective, a mortar with hydraulic and aerial capabilities was created through the incorporation of siliceous and aluminous materials into its composition.

## 3.3.8. Mechanical

The compressive strength values obtained for the mortars produced with OSW replacing up to 20% of the cement weight were slightly lower than those of the reference mortar [53]. The authors commented that the crushed oyster shell particles, because of their fineness, acted to fill the voids of the Portland cement. As the curing time increased, the values increased, and the authors concluded that the requirements of plastering standards were fulfilled.

A study on the influence of different aggregate/binder and water/binder ratios on OSW lime mortars was carried out by Zhang et al. [45]. In terms of compressive strength, an aggregate/binder ratio equal to 2:1 and a water/binder ratio of 0.6 stand out as the composition that offered the best results. After 14 days, these mortars presented a compressive strength of 0.3 MPa, and after 28 days, this value grew to 0.5 MPa. There was a tendency to decrease the resistance with the increase in the amount of water. The addition of glutinous rice was also tested, and an increase in the compressive strength and ductility of OSW lime mortars was observed.

Seo et al. [46], in their investigation, calcined OSW powder at 1000 °C for 3 h and replaced 0, 3, 6, 9, and 12% of cement by weight. The shell comprised 98% pure calcium oxide with a lime crystalline structure. The authors stated that a content of 3% contributed to increased compressive strength. When percentages above 9% were used, the effect on compressive strength was negative. At 3 days after application, the strength of the reference cement mortar showed a value of 34.4 MPa, which was slightly lower than the mortar with 3% of incorporation (36.4 MPa), but higher than the strength of the mortar with incorporation of 12% (22.5 MPa). When considering the mortar at 56 days after application, the strengths for the mortar with 0, 3, and 12% oyster shell incorporation showed 53.7 MPa, 57.5 MPa, and 42 MPa, respectively. Finally, the authors limited the use of OSW to percentages lower than 6% due to the characteristics presented. Through the characterization of the microstructure, the authors defined that the increase in compressive strength in the mortar incorporating 3% calcined oyster shell powder was mainly due to the reduction in the amount of macropores; however, this effect did not stand out in relation to the amount of cement incorporated in the mixtures, considering the other replacement percentages used.

When 8 and 33% of cement were replaced by OSW powders and other marine residues, after 28 days of curing, Ez-zaki et al. [50] found that the compressive strength of the mortars decreased. The authors also indicate that residues and OSW have a slower hydration in relation to cement, which leads to the need for a longer curing time to obtain higher strengths. However, samples with a mixture of OSW and marine residues showed greater resistance than those using only one residue (oyster shell, clam, green mussel, and cockle shell). Another finding was that the heat treatment of calcination at 850 °C was more effective than that at 650 °C for compressive strength, i.e., at 850 °C, the compressive strength was greater. Other authors, such as Ez-zakia et al. [48], reached the same conclusion. However,

the authors commented that despite the observed reductions, the resistance obtained was still among the resistance classes recommended by the standard (42.5 MPa, for example).

The higher flexural (7.55 MPa) and compression strengths (49.2 MPa) were obtained when the amount of cement replaced by OSW powder was 5%, according to the study developed by Bin-Yang et al. [52]. The compressive strength using this replacement percentage was higher for all ages, and at 28 days, the values were 5% higher than the reference mortar. The influence of the type of milling on the resistance was the evaluated factor. The authors reported that the OSW powder with wet grinding was finer and more homogeneous; it also provided a better effect of filling and densifying the structure of the hardened mortar than when using dry grinding. The authors recommended substitutions of 5 and 10%. From these substitutions, the authors reported that for the same water/binder ratio and an increase in the amount of OSW incorporation, the mixture loses workability and gains increased porosity due to the high surface area of the residue.

Liao et al. [51] concluded that as the rate of incorporation of calcined OSW powder increases (0, 5, 8, and 10%), the increases in flexural and compressive strength are clear. The increases are greater for the younger ages, and after 7 days, the process is slower due to a decrease in the hydration rate and pozzolanic reaction. The incorporation of metakaolin as a replacement for 10% of the cement, together with the OSW powder, favored the pozzolanic reactions. The highest flexural and compressive strengths were 10.57 MPa and 54.45 MPa at 90 days, respectively.

Liu et al. [56] concluded that when using different finenesses of calcined OSW powder, improvements in compressive strength were obtained. The initial strength was increased with the mesh size of 6000 OSW powder (3 days), compared to the 1250 and 3000; however, at more advanced ages, the 3000-mesh particle size was more important for the increase in strength (7 and 28 days), presenting a value of 46.5 MPa at 28 days, while the values for the reference cement mortar were 46.2 MPa. In the case of mortars with particles of 1250 and 6000 it was 38.5 and 45.6 MPa, respectively. The authors stated that the increase in compressive strength at more advanced ages was due to the amount and density of the C-S-H gel. According to the authors, the particle size of 10–20  $\mu$ m in the mortar produced with OSW with the mesh particle size of 3000  $\mu$ m can have a "nucleation effect" similar to the cement hydration process. This fact helps to increase the C-S-H content and evenly distribute the hydration products over the entire transition zone of the interface.

Liao et al. [59], when using the maximum diameter for OSW of 0.6 mm as a substitute for river sand with different proportions (0, 10, 20, and 30%), found that the flexural strength increased with the age of curing and amount of sand replacement. With the proportions of 20 and 30% replacement, the flexural strength that was supposed to be reached at 28 days was achieved at 7 days. Regarding the compressive strength, it was increased with replacements greater than 10% compared to the reference mortar. The cement mortar with 30% of replacement, at 28 days, showed the highest compressive strength among the mixtures studied. Resistances still increased after 90 days of curing, indicating an increase in this property in the long term. These strength contributions were due to the filling of voids and finer porous structures caused by the use of OSW residue. The filling effect of OSW obstructed the connection between the pores, resulting in a denser arrangement and contributing to the increase in the strength.

In the case of the study by Liu et al. [49], the authors analyzed cement mortars produced with the replacement of sand by 20% OSW pre-treated with polyvinyl alcohol and sodium silicate and found that the use of the residue decreased the mechanical performance. Furthermore, it is noteworthy that the mortars with crushed aggregates and treated with sodium silicate showed better compressive strength in the early ages (at 3 days, the compressive strength was 16% higher than the reference mortar), while greater flexural strength for all ages was observed (up to 90 days). Thus, the authors concluded that polyvinyl alcohol has a negative effect on the mechanical properties of the mortars. Larger amount of pores of about 0.1  $\mu$ m in diameter were found in these mortars, which might explain the lower resistance. On the other hand, sodium silicate participated in the hydration reactions of the material and improved the mechanical performance.

Chen et al. [44] studied a reference cement mortar with 30% OSW replacing natural sand. Other mortars were produced with OSW in the same proportion, albeit with the incorporation of fly ash and granulated blast-furnace slag in the proportions of 20%, 30%, and 40% as a replacement for cement. At 3 days of age, the reference mortar stood out in terms of flexural strength. At 90 days, the strengths of the other mortars were similar to the reference mortar. In relation to compressive strength, the reference mortar showed 38.56 MPa at 90 days, and the mortars with levels of fly ash and granulated blast furnace slag showed lower strengths. The authors commented that pozzolanic reactions did not compensate hydration products presented in cement replaced by fly ash and ground granulated blast furnace slag. However, at more advanced ages, strengths were increased, approaching the reference mortar, for the replacement mortars, which mainly used ground granulated blast furnace slag due to its higher pozzolanic activity. The reason for the increase in the mechanical properties from 7 to 28 days is the pozzolanic reactions that occurred between fly ash or granulated blast furnace slag and Ca(OH)<sub>2</sub> and generated secondary hydration products. The C-S-H gel enveloped the capillary pores of the mortar with OSW, resulting in a denser microstructure.

Yoon et al. [2] concluded that there was no significant reduction in the compressive strength of cement mortars containing small particles of OSW as a sand substitute. However, after producing mixtures replacing 100% of the coarse (LOS: 4.75–2 mm) and fine (SOS: 2-0.074 mm) sand particles, the authors concluded that the replacement of fine particles led to results closer to those of the reference mortar. The failure planes of mortars containing coarse aggregates were affected by the nature of oyster shell particle formation. The increase in the volume of the oyster shell mixture caused a reduction in the volume of the cement paste, and, consequently, the strength decreases. On the other hand, when there is an increase in the relative density of the oyster shell particles, the contact resistance of the porous structure increases, and, therefore, the compressive strength is high. Some factors are involved in the process of mortars acquiring strength, such as mixing, cement paste hardening, the contact interface between the oyster shell particles, and the solid matrix. From 7 to 28 days, the mortars mixed with OSW experienced a significant increase in compressive strength. Between 28 and 60 days, the increase was smaller. By adding fly ash, an increase in compressive strength results was observed at 60 days. Ethylbenzene was also added to the mixture, though no contribution was observed when the resistance values were evaluated.

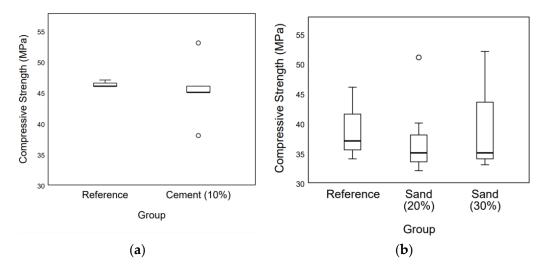
Liao et al. [58] replaced 20% of river sand with OSW in the cement mortar in fine, medium, and coarse dimensions, finding that smaller particles contributed more effectively to the mechanical characteristics. Thus, the flexural and compressive strengths, as well as the elastic modulus, were favored with the increase in the curing period and the decrease in the particle size. According to the authors, coarse aggregates tend to have large cracks and reduced strength, though they still have greater porosity in relation to the other studied particle sizes (fine: 8.74%, medium: 10.26%, and coarse: 10.48%). A smaller particle size made the mortar denser, and its resistance would be greater in relation to the deformation under the action of the same load. It was observed that the strengths grew rapidly in the first ages and then gradually stabilized. The authors also reported an increase when superplasticizers were used.

The study of Wang et al. [60] compared different percentages of sand replacement by OSW and fly ash. The conclusion was that the compressive strength up to 28 days of age was similar for all mixtures. At 120 days, the maximum increase was around 14%, indicating that the replacement of sand by fly ash and OSW sand increased the later strength values of cement mortar. The authors commented on the limitation of using oyster shell in increasing the compressive strength, which, with the use of fly ash, could increase the pozzolanic reaction, improve the compactness of the mortar structure, and increase the bond strength of the aggregate–mortar interface. The OSW was also tested as a powder in the composition of the mortars used to reinforce the brick masonry through the shear test [54]. The authors found an increase in the shear strength of the brick masonry wall when the original mortar was partially replaced by a mortar with OSW powder. They also reported that the mortar produced with OSW was used as reinforcement of ancient masonry in China. Other studies, such as that carried out by Chen et al. [55], concluded that the compressive strength test in brick masonry reinforced with OSW based-mortar fulfilled the design requirements (5.26 MPa).

In the case of the study by Yoon et al. [5], cement mortars were produced with two different ratios (varying the amount of sand). Replacements were up to 80% of the amount of sand per OSW. Up to 20% incorporation of crushed oyster shell was found for the ratio that had the highest amount of sand (cement/mixed soil ratio = 0.1), up to 40% incorporation of crushed oyster shell was found for the ratio that had the least amount of sand, and there were no significant reductions in compressive strength. With 20% and 40% of OSW incorporation, the differences in compressive strength were 0.8 and 0.7 times compared to the reference ratio, respectively.

Furthermore, OSW powder was used in the study by Hong et al. [57] to replace 50% of the natural mortar with a solution with water and OSW powder in its composition (for bioconcrete). The compressive strength results of this mortar indicated higher strengths at 3 and 7 days than the strengths of the reference mortar. The compressive strengths at 3 and 7 days were 74% and 87% of the strength at 28 days, respectively, for mortars produced with the solution. The OSW powder treatment was carried out with nitrate acid to form calcium ions and nitrates. According to the authors, calcium and nitrate ions are harmless to the structure and can have a positive effect on concrete properties. The decrease in the water-binder ratio due to the absorption of the non-acid-treated oyster shell may also have contributed to the increase in the initial compressive strength. At 28 days, it was the reference mortar that showed the highest compressive strength. In this case, ground oyster aggregate may not have the same effect as current fine aggregates used in concrete. The authors emphasize that oyster shells are an ideal source of calcium and have a good cost-benefit ratio that increases bacterial survival and promotes crack sealing. The authors recommended additional testing to understand the effect of acid-treated oyster shell in the development of long-term strength in concrete.

Mortar composites for concrete applications correspond to 79% of the studies found. With this finding, the percentage replacements of cement (10%) and sand (20 and 30%) by OWS that were most repeated were identified and are included Figure 5.



**Figure 5.** Box-plot of compressive strength results: (**a**) reference and 10% replacement of cement [51–53,56]; (**b**) reference and 20% and 30% replacements of sand [5,44,49,58–60].

Box-plot charts are useful in situations involving outliers because they are based on the median of the results. The results of compressive strength of mortars produced with oyster shell can show great variations compared to the results of reference mortars (but even so, they comply with the requirements demanded by the standards). In this case, the mean and the standard deviation may not be good measures. For the composites produced with 10% replacement of cement using OSW powder, two discrepant results are presented (about 38 MPa [56] and 53 MPa [51]), and these results are below and above the lower and upper limits calculated by the box-plot, respectively (Figure 5a). The composites produced with the replacement of 20% of sand by OSW showed a discrepant result (about 51 MPa [59]) above the upper limit calculated via the box-plot. No discrepant results were found in the compressive strength of reference composites and with the replacement of 30% of sand by OSW (Figure 5b).

#### 3.3.9. Shrinkage (Autogenous and Drying)

The autogenous shrinkage was analyzed by Seo et al. [46]. The authors used  $100 \times 100 \times 400$  mm samples and a data logger to record the behavior of autogenous shrinkage until changes in shrinkage reached a constant result. The results of maximum initial expansion of cement mortars incorporating OSW powder calcined in percentages of 3, 6, 9, and 12% were +50, +55, +99, and +155 microstrains, respectively. These expansions were greater than those of the mortar produced only with cement. As the OSW powder has CaO as its main compound, as the hydration process progressed, Ca(OH)<sub>2</sub> was produced, and the material was subjected to the so-called chemical shrinkage. As time went by, the hydration of the calcined OSW mortar decreased, and the factor responsible for the hydration in later ages was the cement clinker. However, the authors reported that this initial expansion of the matrix in the initial stage of hydration was compensated via autogenous shrinkage (the results of the autogenous shrinkage of the mortars were -520, -432, -527, -393, and -422 microstrains for incorporations of 0, 3, 6, 9, and 12% calcined oyster shell powder samples, respectively, after 25 days).

The drying shrinkage of the mortar was tested in four studies. Liao et al. [51] and Lertwattanaruk et al. [53] tested drying shrinkage in accordance with ASTM C596, and Liao et al. [58] and Wang et al. [60] used ASTM C157/157M and ASTM C827, respectively.

Liao et al. [51] commented that the drying shrinkage increased in relation to the curing time. A percentage replacement of cement by OSW powder of around 8% seemed to be ideal. At 90 days, when using a total of 10% OSW powder, the shrinkage value was already 25% greater than that of the mortar with 8%. Drying shrinkage was influenced by the loss of water from the pores of the mortar. A lot of water is spent in the hydration process, and the same amount of water was used in all mixtures. Higher drying shrinkage, with higher OSW powder incorporations, was found due to the large pore water consumption that prevented the pozzolanic reaction sequence.

Lertwattanaruk et al. [53] found that ground OSW (13.93  $\mu$ m) had greater fineness compared to Portland cement (22.82  $\mu$ m), and the mortars produced with these shells showed lower shrinkage than the reference mortar. The authors explained that the incorporation of shells caused a refinement of the pore structure due to the segmentation of large pores. This outcome made the internal structure denser, with fewer voids and less shrinkage.

Over 90 days, Liao et al. [58] observed that it was the aggregate with the fine granulometric range that presented greater shrinkage via drying in relation to the coarse aggregate. The results reached values greater than the limits of norms (0.075%). Fine particles generated a more compact structure, and a greater magnitude of capillary pores tend to form greater shrinkage in the mortar. The authors found that by using a superplasticizer additive in the composition of the cement mortar with oyster shell residue, it was possible to reduce the shrinkage rate. Wang et al. [60] observed that when using low replacement percentages of fly ash and OSW sand, shrinkage rates decreased (with 5% replacement it was 0.045%) compared to the reference mortar (0.065%). According to the authors, this reduction in the traction rate was reduced because the cement paste completely enveloped the aggregate, and the C–S–H pozzolanic reaction hydrate gel completely filled the pores, making the structure more compact.

#### 3.3.10. Water Absorption

Cement mortars produced with OSW powder showed low water absorption coefficients, decreasing with a rise in the curing time, when compared to the reference mortar [51]. As the OSW powder content increased, the value of the water absorption coefficients decreased. The authors concluded that the decrease in porosity is the main reason for the reduction in the rate of water absorption.

In the study developed by Chen et al. [44], the mortars that contained only 30% OSW showed lower absorption values for all ages evaluated (7, 28, and 90 days) than those that had cement replacements by supplementary cementitious materials (fly ash and granulated slag of blast furnace). As the curing time increased, the mortars with the supplementary cementitious materials showed greater decreases in water absorption than the reference mortar. This finding indicated that, at an early stage, the difficulty of forming the C-S-H gel necessary to have a denser microstructure was great, as well as the fact that at later ages, its contributed to pozzolanic reactions, through the use of supplementary cementitious materials, in place of cement.

Wang et al. [60] concluded that cement mortars with substitutions of up to 30% of sand by OSW became more compact, and, consequently, the absorption rate reduced. The authors commented that, as the curing time increased, the pozzolanic reactions of the fly ash occurred, producing C-S-H gel, filling the pores, and decreasing the absorption rate. The reductions were from 1.4 to 6.5% for mortars with 10 and 30% replacement, respectively, compared to the reference mortar.

Liu et al. [49] found that as the curing time increased, the water absorption of mortars containing 20% of oyster shell aggregate treated with polyvinyl alcohol or sodium silicate decreased in relation to the reference mortar. Furthermore, the use of polyvinyl alcohol in the pre-treatment appeared to be more effective than using sodium silicate to reduce water absorption at 28 and 90 days. A greater reduction in the water absorption with sodium silicate mortar can be attributed to the reference mortar. Moreover, when the mortars contained a solution with polyvinyl alcohol, a thin film on the surface of the crushed oyster shell was formed without changing its density.

Liu et al. [56] found that capillary water flow decreased when the mesh size of calcined OSW powder increased. The authors produced cement mortar with supplementary cementitious materials and concluded that the pore structure of the mortar became more compact due to the larger size of OSW powder (mesh size of 6000) and its voids being filled by the microaggregates (without secondary hydration).

#### 3.3.11. Volume of Permeable Void

Considering the cement mortars studied by Liao et al. [59] and the maximum particle size of OSW of 0.6 mm, all permeable void volume results became smaller as a greater amount of OSW powder residue was incorporated. As the curing time increased, the values also decreased. This consequence is explained by the fact that there is a gradual hydration, leading to a complete hydration.

Liu et al. [49] found that cement mortars with treated OSW aggregates had lower permeable void volume values. The lowest permeable void volume was found when the OSW aggregate was treated with sodium silicate. The addition of OSW aggregates treated with polyvinyl alcohol predominantly left apparent density unaltered, compared to the reference mortar. The decrease in apparent density and, consequently, the increase in apparent porosity with the incorporation of calcined OSW and other marine residues were reported in the studies by Ez-zaki et al. [48] and Ez-zaki et al. [50]. The authors attributed this characteristic to the greater volumes of water that cement mortars using high amounts of residues needed to reach an adequate consistency. The mixtures had the same water/binder ratio, which made it more difficult to mold these mortars, causing greater formation of air bubbles in the material. One factor influencing a greater water demand was the fact that sediments presented a greater Blaine surface area compared to cement.

The study by Liao et al. [51] found that the apparent density was increased and the permeable void volume was decreased as the incorporation content of OSW powder increased (0, 5, 8 and 10%). The authors commented that the products of the pozzolanic reaction reduced the total volume of pores. The Ca(OH)<sub>2</sub> that did not participate in the pozzolanic reaction was incorporated into the hydration products, which made the cement mortar structure more compact.

#### 3.3.12. Permeability Coefficient (Water and Gas)

Liao et al. [59] achieved water permeability coefficient values for reference cement mortar that were 75% higher than for mortar produced with 30% replacement of river sand by OSW powder. As the percentage of replacement increased, the water permeability coefficient of the mortars decreased. Nonetheless, from 28 to 90 days, the coefficients both decreased. When 30% of river sand was replaced by OSW powder, this decrease was 18%. The authors mentioned that this effect was caused by the particle diameter of OSW powder being smaller than cement. In this case, the initial pores were filled and led to a decrease in the proportional water permeability coefficient, as well as in smaller capillaries found for mortars with OSW powder, resulting in a denser structure.

Liu et al. [49] emphasized that cement mortars with OSW aggregates had a high capillarity coefficient, which could be compensated via surface treatment. At 28 and 90 days, the water permeability results of the mortars containing polyvinyl alcohol and sodium silicate decreased both in relation to the reference mortar and over time. The reduction was greater for the mortars containing sodium silicate due to their higher degree of hydration in relation to the other mortars.

It was possible to verify, in the study by Chen et al. [44], an increase in the water permeability of OSW mortars, in which natural sand was replaced by OSW and cement replaced by fly ash and ground granulated blast furnace slag. However, at older ages, this impact was reduced. This outcome was due to pozzolanic reactions and curing making the material more impervious to water infiltration with a more compact structure.

Ez-zakia et al. [48] reported that when 33% of cement was replaced by marine residues and calcined OSW, a reduction in the connection of the internal pore structure and, consequently, an improvement in the apparent gas permeability compared to the reference mortar were verified.

Liao et al. [51] concluded that the water permeability coefficient has a maximum reduction of up to 36.19% in 28 days, as well as 29.28% in 90 days, when the replacement of cement by OSW powder was 10%. The reduction values were greater than the ones of the reference mortar for all ages. The reduction caused by OSW powder had a greater effect in the early curing ages. The authors commented on the blockage in the diffusion and refinement pathways of the pore structure, with both caused by the products of the hydration reaction and the pozzolanic reaction.

## 3.3.13. Microstructure

According to the results found by the authors Seo et al. [46], the different contents of calcined OSW powder (3, 6, 9 and 12%) that replaced the binder in cement mortars did not induce differences in relation to the formation of micropores. On the other hand, as the amount of incorporation of calcined OSW powder increased, smaller amounts of pores were found, with sizes ranging from 100 to 1000  $\mu$ m (macropores). Overall, all

samples showed components due to the presence of hydration and carbonation products, as well as unhydrated clinker minerals (i.e., alite, belite, portlandite, ettringite, and calcite). The presence of CaO was not identified in all pastes, indicating that the incorporated calcined OSW powder was mainly consumed during hydration. In the case of pastes with higher amounts of calcined OSW powder, X-ray diffraction (XRD) patterns revealed the coexistence of portlandite with calcite. This fact was due to portlandite being produced at early and additional stages, albeit with a certain amount of portlandite consumed by natural carbonation.

Through XRD analysis, the authors Liu et al. [49] reported that the mortars at 28 days did not show notable changes in the chemistry of the mixture. The sodium silicate treatment did not demonstrate any geopolymeric-binding phase or alkali-activated material in the spectra in the Fourier transformed infra-red spectroscopy (FTIR) tests. Sodium silicate acted as an active addition to the mortar by reacting with CH and forming CSH gels. This reaction accelerated the hydration process and contributed to a more advanced hydration in the cement paste. The scanning electron microscopy (SEM) showed that the transition zone of the mortar with aggregate treated with sodium silicate proved to be less porous, which must have arisen as a result of greater and better hydration providing a denser structure. However, more pores were found in the cement paste. Nonetheless, via SEM, the mortars containing the treated OSW aggregates were more homogeneous and denser than the reference mortar.

Chen et al. [44] studied cement mortars with 30% OSW (maximum diameter of 5 mm) replacing natural sand, as well as with the incorporation of fly ash and ground granulated blast furnace slag in the proportions of 20%, 30%, and 40% replacing cement. In terms of microstructure, OSW particles randomly distributed throughout the internal structure of the mortars were observed. When observing the mortars with OSW at 7 days via energy-dispersive X-ray spectroscopy (EDS), it was possible to observe high peaks of Ca and Si. By analyzing the chemical compounds of the microregion, the formation of C–S–H gel in the hardened mortar over time was confirmed. At initial ages (7 days) with the incorporation of fly ash and ground granulated blast furnace slag, low degrees of hydration were indicated. On the other hand, from 7 to 28 days, the pozzolanic reactions took place between fly ash or ground granulated blast furnace slag and Ca(OH)<sub>2</sub>, forming secondary hydration products. A significant amount of C-S-H gel was generated involving the capillary pores, resulting in a denser microstructure of the hardened OSW mortars.

Ez-zaki1 et al. [50] report that the analysis of cement mortar specimens via EDS found hydrated products (portlandite, ettringite, monosulfoaluminate), anhydrous phases of cement ( $C_3S$ ,  $C_2S$ ,  $C_4AF$ ), and other compounds, such as calcined OSW powder and marine residues in the form of SiO<sub>2</sub> and CaCO<sub>3</sub>. The morphologies and chemical compositions of the different mixtures, identified at 8 and 33% and by varying the sediments, were similar to the reference paste produced only with cement.

The OSW lime XRD test results were discussed by Zhang et al. [45]. It was possible to observe different amounts of  $\beta$ -calcium silicate ( $\beta$ -CS), with the calcinations carried out at temperatures of 800, 1000, and 1200 °C in OSW lime. The authors commented that the silicate presented in the clay reacted with the calcium of the shells during the calcination and formed  $\beta$ -CS. The amount of  $\beta$ -CS and the degree of calcium carbonate reaction were influenced by the calcination temperature. Calcination at 1000 °C disintegrated almost all of the calcium carbonate and produced a greater amount of  $\beta$ -CS, which is beneficial for the durability of OSW lime mortars.

Bin-Yang et al. [52] confirmed, through EDS analysis, that the main product of hydration of cement  $Ca(OH)_2$  and  $SiO_2$  and OSW is  $CaCO_3$ . The authors reported that the results of the XRD analysis did not identify chemical reactions between the cement and OSW powder, i.e., the residue was not calcined, had the effect of filling in the mortar, and did not affect the stability of the cement.

Liao et al. [51] did not observe significant changes in the type of chemical compound of the mortar with substitutions of different contents of calcined OSW powder with cement. However, an existing peak of CSH generated from the increase in the pozzolanic reaction was observed due to the use of OSW powder. The amount of CSH increased as the percentage of shell replacement (0, 5, 8 and 10%) increased.

Liu et al. [56] verified, through the use of SEM, that the incorporation of calcined OSW powder considerably improved the amount of CH in the mixture. The formation of CH provided a high alkalinity environment for the system to promote secondary hydration of lithium slag and ground granulated blast furnace slag. However, when the OSW size was too large, it was promising to secondary hydration of supplementary cementitious materials, though the filling effect improved the mortar's waterproofing.

## 3.3.14. Chloride Ions Diffusion Coefficient

An improvement in chloride ion diffusion resistance was found in cement mortars with greater replacements of sand by OSW powder in the study by Liao et al. [59]. At 28 days, the mortar with 30% replacement of sand by OSW powder showed the lowest value  $(3.25 \times 10^{-12} \text{ m}^2/\text{s})$ , which was almost 75% of the chloride ion diffusion coefficient of the reference mortar  $(4.35 \times 10^{-12} \text{ m}^2/\text{s})$ . The internal structure of finer pores, which were denser and with lower permeability, made it difficult to transport chloride ions. The authors also reported that this result was mainly obtained via the penetration of chloride ions through the permeable pores that hardened the mixture (the more chloride ions penetrate into the cement paste, the more reactions with ions, such as Ca<sup>2+</sup> and Al<sup>3+</sup>, can occur in the pores and form salt of Friedel, resulting in the reduction in pores). The authors mentioned that the use of metakaolin in the composition of the mortars contributed to the densification and, consequently, the resistance to the diffusion of chloride ions, due to the pozzolanic reaction and generation of calcium carboaluminate. Finally, the authors emphasized that mortars with OSW powder reduced the risks of corrosion of steel in marine environments, increasing the durability of the mortars.

Considering the ages tested (28 and 90 days), Liu et al. [49] found that cement mortars containing treated OSW aggregates performed better in terms of resistance to chloride ion penetration, compared to the reference mortar. The values decreased with the advancing age of the cure. The authors commented that sodium silicate accelerated the hydration reaction, forming more gels that filled the pores and decreased the penetration of chloride ions.

By determining the penetration coefficient of chloride ions via a rapid chloride ion permeability test (RCPT), it was noticed that the results were influenced by the contents of the materials used and the curing time. In this case, the fly ash particles and ground blast furnace granulated slag had greater ability to adsorb and bind the chloride ion, when compared to cement, and with increasing curing time, the penetration of chloride ions inside the mortars with 30% OSW may decrease [44].

Liao et al. [51] stated that a content above 8% replacement of cement by OSW powder did not have a relevant effect on the chloride ions diffusion coefficient. The values found in the tests were lower as the incorporation of residue and the curing time increase. According to those authors, a decrease in porosity and a more compact structure make it difficult to diffuse of chloride ions inside the mortar.

Liu et al. [56], when evaluating the rapid penetration by chloride ions, concluded that as the size of the mesh (in which the calcined OSW was sieved) was reduced, for its replacement as cement, the better was the resistance to the penetration of chloride ions. The authors reported that the effect of filling the non-hydrated particles prevailed, leaving the mortar with a denser structure.

According to Wang et al. [60], the oyster shell is a coastal material and, therefore, has a high concentration of chlorides. Nonetheless, the authors found that despite the chloride ion content of the reference mortar  $(0.028 \text{ kg/m}^3)$  being lower than the cement mortars

with replacements, the value measured with the 30% replacement of the sand by OSW  $(0.146 \text{ kg/m}^3)$  met the specification  $(0.3 \text{ kg/m}^3)$ .

#### 3.3.15. Eco-Efficiency

The authors Liao et al. [59] discussed some ways to promote sustainability in engineering through the reduction in environmental impact during the maintenance and work process, and the increase in durability with the best performance. The authors found that with 10 and 30% replacements of river sand by OSW powder, more sustainable and better performing mortars can be obtained. The results showed that the greater the amount of replacement of river sand by OSW powder, the lower the total CO<sub>2</sub> emissions (per m<sup>3</sup>) in relation to the reference cement mortar. By using this material, it was possible to reduce total CO<sub>2</sub> emissions and the area occupied by oyster shell waste and environmental pollution.

Liao et al. [58], with the replacement of 20% of river sand by fine OSW, reported the contributions of OSW use in terms of reducing river sand demand, as well as ensuring proper disposal of OSW through sustainable recycling and the production of ecological and high performance mortars. The authors recommended the production of cement mortars with fine OSW, resulting in an inexpensive and ecologically beneficial material, with good workability and mechanical properties. Thus, the studies by Zhang et al. [45] also highlighted the advantages of using shell lime in the construction industry through OSW calcination, in view of its use for the restoration of old buildings, replacing gypsum in decorations and as a binding material in building houses, being both beneficial for the construction industry and the environment. Both authors [45,58] commented on environmental concerns in China, which are linked to the illegal dumping of shells due to people's lack of awareness of environmental protection and lack of technical support. Zhang et al. (2019) also mentioned on the possibility of collecting and storing CO<sub>2</sub>, generating carbon capture and an energy reserve, with both methods produced during the calcination of shells.

In the study by Liao et al. [51], the mortars that experienced the replacement of cement by 5%, 8%, and 10% of the calcined OSW powder had reductions of 9.1%, 13.6%, and 13.6% in relative emissions of  $CO_2/MPa$ , respectively, and reductions of 7.5%, 11.3%, and 13.2% in relative costs (per MPa), both in relation to compressive strength and compared to mortar with 0% OSW powder. Thus, the authors highlighted the benefits of economic and environmental cost reduction generated via the production of sustainable and high performance mortars with OSW. The authors still hope that the use of OSW will be adopted on a large scale in the construction industry in the future.

Finally, Chen, et al. [44] evaluated the  $CO_2$  emissions in mortars produced with fly ash and granulated blast furnace slag (both as a binder) and using OSW (as an aggregate). The authors found that the use of these residues would compensate the demand for natural fine aggregate and cement, reducing  $CO_2$  emissions and associated costs. When 30% of the aggregate was replaced by OSW, and 40% of the cement was replaced by fly ash and granulated blast furnace slag, the greatest reductions (38.74% and 38.39%) in total  $CO_2$  emissions occurred. A greener mortar with adequate engineering properties could be produced by incorporating fly ash and granulated blast furnace slag and OSW in limited quantities.

## 4. Discussion

Table 6 summarizes the results obtained in the investigations analyzed in this work, namely the properties of the oyster shell waste, the general results, the behavior of the composites when using other materials, and the percentage replacement of traditional materials in composites by oyster shell.

Properties	General Results (Compared to the Reference Composites)	Use of Other Materials	Recommendations and Comments
Workability	Calcined OSW Powder: lower or equal; OSW Powder: higher; OSW sand: lower.	The amount of water consumed by oyster shells is greater than the figures for other types of shells. In this case, a superplasticizer additive can help to minimize water usage.	
Heat of hydra- tion/Setting time	Calcined OSW Powder: higher heat of hydration; OSW Powder: higher setting time; OSW sand (with coarse particle size): higher setting time	In combination with other materials (metakaolin, gismondine), it shows good hydraulic reactivity. The use of fly ash can help to increase the setting time. With the introduction of siliceous and aluminous materials, mortars with both hydraulic and aerial capabilities are produced.	Replacements cement in the range of 8% to 16% shows good hydraulic reactivity. The longer setting time is obtained when 30% of sand is replaced.
Mechanical strength	Calcined OSW Powder: lower and higher; OSW Powder: lower; OSW sand: lower and higher; Solution: higher (for 3–7 days).	Composites with OSW sand and other marine waste show higher strengths than those with only one waste material. The use of metakaolin favors pozzolanic reactions. Fly ash contributes to pozzolanic reactions (higher strengths at later ages). The use of superplasticizer increases strength. Pre-treated shells decrease strength. The pozzolanic reactions obtained with the use of blast furnace slag and fly ash do not compensate for the hydration products of cement. WOS powder treated with nitrate acid is harmless to the structure.	The suggested cement substitutions are 5% and 10%, as well as being lower than 6%. Thermal treatment of calcination above 850 °C is more effective. Replacement with 30% or 20% and 40% of sand show the highest strengths.
Shrinkage (autogenous and drying)	Calcined OSW Powder: lower and higher; OSW Powder: lower; OSW sand: lower and higher (with fine particle size).	The use of a superplasticizer additive in composites with OSW sand reduces the shrinkage rate. Composites with fly ash replacement percentages also decrease shrinkage.	A percentage of about 8% of cement replacement is ideal.
Water absorption	Calcined OSW Powder: lower; OSW sand: lower.	Composites with supplementary cementitious materials (fly ash and ground granulated blast furnace slag) show greater reductions in water absorption. Treatment of the shell with polyvinyl alcohol or sodium silicate results in lower water absorption.	Replacement of 20% and 30% of sand has lower absorption results.
Volume of permeable void	Calcined OSW Powder: lower and higher; OSW sand: lower and higher.	When using calcined OSW powder and other marine waste, porosity increases. The porosity of the shell decreases with pre-treatment with sodium silicate. Incorporation of pozzolanic materials (metakaolin) contributes to a reduction in the total volume of pores.	-
Permeability coefficient	Calcined OSW Powder: lower; OSW sand: lower and higher.	The use of treated OSW aggregates (polyvinyl alcohol and sodium silicate) decreases permeability. The use of other materials (fly ash and slag) presents higher permeabilities.	Replacements of 10% and 33% of cement and 30% replacement of sand show lower permeability coefficients.

 Table 6. Results obtained via comparative analysis with OSW incorporation.

Properties	General Results (Compared to the Reference Composites)	Use of Other Materials	Recommendations and Comments
Microstructure	-	Treating the shell with sodium silicate makes the mixture more homogeneous and dense. The use of ground fly ash or ground granulated blast furnace slag generates a denser microstructure (at higher ages).	Calcined OSW Powder: equal micropores and decreased macropores. OSW Powder: filling effect does not affect cement stability. OSW sand: distributed throughout the internal structure of the mortar.
Diffusion coefficient of chloride ions	Calcined OSW Powder: lower; OSW sand: lower and higher.	The use of metakaolin increases the resistance to chloride ion diffusion. Treated shells reduce chloride ion penetration. Chloride ion penetration decreases over time with the use of fly ash and granulated blast furnace slag.	A cement content replacement above 8% by WOS powder does not have a relevant effect on the chloride ion diffusion coefficient. Substitution of 30% aggregate by OSW either presents lower results or meets the standards.
Eco-efficiency	Calcined OSW Powder: reduce CO <sub>2</sub> emissions; OSW sand: reduce CO <sub>2</sub> emissions.	More sustainable composites with adequate engineering properties can be produced by incorporating fly ash and granulated blast furnace slag (as a replacement for cement) and limited amounts of WOS sand.	With replacements of 10% and 30% of river sand with WOS powder, more sustainable and better-performing mortars can be obtained.

Table 6. Cont.

Based on reference composites, the authors found some causes of the behaviors presented in the composites that involved the substitution of traditional materials by OSW.

When the calcined OSW powder replaced the binder, larger amounts of water were required for the formation of  $Ca(OH)_2$ . In the case of the powder used without calcining, the mixture had smaller amounts of cementitious material and lower water retention. For this reason, the mortars produced required smaller amounts of water. As the OSW sand had higher water absorption compared to the water absorption capabilities of the natural aggregate, the composites produced with the residue had lower workability. The water demand in the mixtures increased as the fineness of the OSW aggregate increases.

The reactions that occurred with calcined OSW powder generated higher heat of hydration compared to the reactions of cement in the early stages. The combination of calcined OSW powder with other materials (e.g.,: metakaolin, gismondine) and the high fineness of these materials caused a large calorimetric effect. The replacement of cement by calcined OSW powder in the range of 8 to 16% was represented by the percentages with the best hydraulic reactivity [48]. Replacing cement with powder without calcining made the hardening process slower, which could be interesting for plaster application in hot climates. In the case of aggregate replacement by OSW, the longest setting time was achieved when 30% was incorporated [59]. The fly ash reduced the reactions that cause prolonged setting times.

The results of the mechanical properties increased due to the reduction in the number of macro-pores when the cement was replaced by calcined OSW powder. The use of noncalcined OSW powder and other marine debris decreased the strength because they had a slower hydration compared to the composites produced with cement, which led to the need for a longer curing time to be implemented to obtain higher strengths. Mechanical strength increased with curing age when OSW sand, as the aggregate, was used due to the filling of voids and finer porous structures (the finer particle size of OSW aggregate generates higher mechanical characteristics). The powder obtained through wet grinding could achieve higher fineness and homogeneity, as well as improve the filling effect. In this process, the composites present higher mechanical strength results. The incorporation of metakaolin with calcined OSW powder in composites as a cement replacement favored pozzolanic reactions. Fly ash contributed to the pozzolanic reactions of the composites with OSW sand (increased strength at higher ages). However, studies also report that the pozzolanic reactions obtained through the replacement of cement with granulated blast furnace slag and fly ash do not compensate for hydration products present in the cement in composites produced with OSW sand. Although some composites had lower strengths than the reference compositions, the results obtained were within the parameters of the standards (coating, laying and mortars, and mortar for concrete). The strength found in composites with OSW powder (without calcined) presented lower results. However, the ground oyster shell particles, being finer, filled the voids of Portland cement. With the increase in curing time, the values also increased.

The drying shrinkage was influenced by the water loss from the mortar pores. Higher drying shrinkage with higher substitutions of calcined OSW powder was found due to the large pore water consumption. As the hydration process progressed, the CaO compound produced Ca(OH)<sub>2</sub>, and the material possessed higher chemical shrinkage. Composites of OSW sand with smaller particle sizes obtained higher shrinkage due to higher capillary pore formation. In other studies, OSW powder (without calcined) produced a composite with denser structure and fewer voids, decreasing shrinkage. In the case of the use of OSW sand, the composite that obtained the least shrinkage involved the use of fly ash in the mixture.

The decrease in porosity is the main reason for the reduction in water absorption rates in composites with cement replacement by calcined OSW powder. The smaller the particles of the calcined OSW powder possess, the lower the water absorption. The pozzolanic reactions of the supplementary cementitious materials also contributed to filling the pores and decreasing the absorption rate (for advanced ages). The composites with treated OSW sand presented lower water absorption in the mortars.

The treatment of OSW sand also resulted in lower permeable void volumes in the composites. However, the composites that used OSW aggregate and the same water/binder as the reference were more difficult to mold, generating higher porosity. The use of the calcined OSW powder with the mixture of other marine waste also increased the porosity. The hydration of the cementitious materials in the OSW calcined powder generated a lower volume of permeable voids.

The hydration reactions of the cementitious materials of calcined OSW powder also contributed to the decrease in the permeability coefficient (water and gas). The pozzolanic reactions due to the presence of metakaolin decreased the coefficient of permeability. The calcined OSW powder reduced the internal pore structure connection and improved the permeability performance. Using OSW sand particles with smaller particle sizes than cement, permeability decreased due to pore filling and lower concentrations of capillary pores. The treatment (with sodium silicate) of the OSW sand decreased the porosity of the composites due to the higher degree of hydration of the reactive materials. The composites that used pozzolanic materials (fly ash and granulated blast furnace slag) did not decrease the permeability when used in composites produced with OSW sand.

Regarding microstructure, it was observed that the calcined OSW powder did not change the amount of micropores, instead decreasing the number of macro-pores (100 to 1000  $\mu$ m) in the composites. The calcined OSW powder was mainly consumed during hydration, but was also consumed via carbonation. The morphologies and chemical compositions of the reference mixture and the mixture with binder replacement by OSW calcined powder were similar. The amounts of  $\beta$ -calcium silicate ( $\beta$ -CS) varied based on the calcination temperature used for composites produced with calcined OSW powder. The amount of CSH increased as the percentage of cement replacement with calcined OSW powder increased. OSW powder (without calcinate) has a filler effect and does not affect the stability of the cement. OSW sand was distributed throughout the internal structure of the mortar.

The smaller the sieving mesh size of the calcined OSW powder, the higher the resistance to chloride ion penetration. The use of metakaolin in composites produced with calcined OSW powder contributed to densification and, consequently, to the resistance to chloride ion diffusion, due to the pozzolanic reaction and generation of calcium carboaluminate. The composites treated with OSW aggregate showed better performance, while the sodium silicate accelerated the hydration reaction, forming more gels that filled the pores and decreased the penetration of chloride ions. The use of fly ash and granulated blast furnace slag could decrease the penetration of chloride ions over time in composites with OSW sand. Even though the reference composite presented lower ion content than the mortars with substitutions, the result measured using the 30% substitution of sand for OSW met the specification. Finally, the authors emphasized that mortars with OSW powder reduced the risks of steel corrosion in marine environments, increasing the durability of the mortars.

In terms of eco-efficiency, the calcined OSW powder and the OSW sand produced composites with lower  $CO_2$  emissions, while using OSW, it is possible to recycle it and produce sustainable and high-performance mortars. The studies recommend the production of mortars with OSW fine sand (powder), resulting in an inexpensive, environmentally friendly material with good workability and mechanical properties.

Overall, the commented characteristics intensified as the percentages of substitutions were higher, and the results obtained fulfilled the normative requirements.

#### 5. Further Studies

The usage of shell by-products as binder/filler and aggregate in mortar composition can enhance several properties of this material. It is probable that granular materials can be replaced by oyster shell up to 20% with the consent of buyers [5]. The replacement of river sand by oyster shell powder up to 30% is environmentally and economically feasible [44,58,60]. Nevertheless, prior to oyster shell's practical application, it is crucial to evaluate several important aspects of its properties. Given this fact, directions for future investigations are recommended.

Regarding the oyster shell characterization, considering that shells are biological materials whose characteristics can be affected by environmental conditions, future research is required to specify the exposure conditions of the shells and the effects of different cleaning and heat treatment methods, as well as of the storage time. It is critical to identify and investigate the type and influence of the species, the origin of the waste collection (e.g., oyster farms or restaurants), and whether changes in shell behavior are seen, depending on the geographic region of waste collection. Accurate identification and classification of the physical properties of oyster shell dust and aggregates (density and water absorption), as well as the mechanical properties of oyster shell aggregates (toughness, strength, hardness, among others), is important.

The production methodology and workable time of the composites should be further explored. No reports were identified that discussed how the materials were introduced into the mixture and the time the composite required to be fully mixed. These data are even more important when additives are used in the composition of the composites (for example, superplasticizers or air incorporators). In addition to the analysis of the setting time, other tests in the fresh state can be performed. To the authors' knowledge, there are no studies available on rheology. Such tests can help to explain the behavior of composites produced with oyster shell.

Concerning the behavior of oyster shell coating mortars, while some results are available in the literature that discuss the hardened state, more investigations are needed on shrinkage and expansion, porosity, and water transport. Most studies discuss flexural and compressive strength up to 28 days for the coatings. It is important to analyze paste–aggregate bonding, modulus of elasticity, ultrasonic pulse velocity, high temperature resistance, freeze–thaw resistance, color change, and odor of oyster shell coating mortars. In the case of interior wall coverings, the absorption of noxious gases and the adjustment of air humidity are also relevant parameters that need to be analyzed. All of these characteristics, combined with the time factor, need to be checked and compared to conventional wall coverings.

Prototype walls built with mortar coatings using oyster shell in the composition can be tested to discover their impact resistance.

The production of coating mortars with insulating aggregates and oyster shell aggregate should be investigated. Thermal insulation and sound absorption are requirements increasingly demanded in building performance legal standards.

The application of mortars produced from oyster shell for coating and brick laying in new buildings can be further explored. This application extends to old buildings that require the production of rehabilitation mortars. The incorporation of shells in lime mortar production was previously employed hundreds of years ago in coastal regions [61].

The Life Cycle Assessment (LCA) of coating mortars produced using oyster shell powder or aggregate should be studied. The acquisition of the residue (distance from waste collection to the factory), the acquisition of suitable particle sizes (in the form of powder or aggregate), the different substitution percentages used in the mixtures, and the calcination process of the oyster shell should be considered. The LCA obtained should be compared to the LCA of the conventional coating mortar production. These results are important and leave clues about the eco-efficiency of a construction product produced using this marine waste.

Current research, with the collaboration of and financial support from local authorities and associated industries, is essential to boost the industrial application and high-value utilization of these materials, thus promoting the growth of the blue circular economy.

## 6. Conclusions

This paper carried out a systematic review of the scientific production of oyster shellbased composites for use in construction. Most studies were from China, and there is a lack of research from some parts of Europe. In the Americas, Africa and Oceania, no studies were found.

Most studies contemplate strong cement mortars; this approach may have been common due to most studies being focused on oyster shell's use as concrete (79%). Only 21% of the analyzed articles were about coating and laying mortars (lime mortars), demonstrating that there is no equivalent body of research regarding the use of mortars for these applications. As for the thematic lines, most studies addressed issues related to mechanical properties (95%).

OSW was used in composites as powder, calcined powder, sand, and as a solution to replace water. Comparing reference compositions with compositions containing OSW, the following conclusions can be drawn

- OSW Powder: the workability was higher, consequently reducing the setting time, mechanical strength, and drying shrinkage. An increase in mechanical strength was also observed with longer curing time. The powder had a filling effect and did not change the stability of the cement.
- Calcined OSW Powder: the calcination of the powder produced composites with the same workability, which were lower due to the increase in the heat of hydration. Properties such as mechanical strength, shrinkage (autogenous and drying), water absorption, permeable void volume, permeability (water and gas), diffusion coefficient of chloride ions, the amount of macropores, and CO<sub>2</sub> emissions were lower. Other studies showed higher mechanical strength, shrinkage, and permeable void volume. The amount of micropores did not change.
- OSW Sand: workability, mechanical strength, drying shrinkage, water absorption, permeable void volume, water permeability, diffusion coefficient of chloride ions, and CO<sub>2</sub> emissions decreased. On the other hand, studies also demonstrated an increase in the properties of mechanical strength, permeable void volume, water permeability, and diffusion coefficient of chloride ions. In the microscopic analysis, OSW sand was distributed throughout the internal structure of the mortar. The setting time increased

with the increase in the aggregate particle size, and drying shrinkage increased with the increase in finer aggregate particle sizes.

• Sources of nutrients: the compressive strength was higher at 3 and 7 days than the reference composition. As a good source of calcium and cost-effective, oyster shell increases bacterial survival and crack sealing.

In future investigations, it will be possible to cover practical and contextual gaps regarding the use of oyster shell waste (OSW) as powder and aggregate in the production of rendering and masonry mortars, as well as in the rehabilitation of old buildings. The utilization of OSW in other types of composites can also contribute to a better utilization of oyster shell waste and its insertion in the blue circular economy. However, improvements in terms of oyster shell characterization (origin, species, exposure conditions, cleaning and thermal treatment methods, and physical and mechanical properties), mixing methods, and deepening knowledge and testing techniques in the fresh state of the composites can minimize the rejection of this waste's incorporation into building materials. This knowledge of the behavior, durability, and life cycle of the materials of composites produced with aquaculture waste may encourage academia, the construction industry, and local authorities to invest in products manufactured using this technology.

The present research contributes to discussion regarding the potential use of oyster shell waste in the creation of construction composites, as well as its inclusion in the blue circular chain as an alternative material in constructive solutions.

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