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Recycling Mussel Shells as Secondary Sources in Green Construction Materials: A Preliminary Assessment

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Abstract: This paper reports the development of novel green bio-composite mortars obtained by reusing mussel shells, a waste from the fish canning industry, as recycled aggregate, used for the first time in total substitution to the traditional sand. It suggests that this is a valid alternative to their usual disposal in landfills because the organic matter is potentially dangerous to humans and the environment. Different waste-based cementitious mixes were tested and compared to a traditional OPC mortar. The manufacturing process was performed at ambient conditions (20 °C, 65% RH) with highly sustainable results and consisted of simple operative steps reproducible in a real building site. The engineering performance was investigated to preliminarily assess the novel material potentials in construction. The main results showed that recycling mussel shells as aggregate while considerably decreasing the mechanical resistance (up to 60% in bending and 50% in compression), mixes could still find proper building applications (either structural, light partition, and plastering) according to the relevant standards. Moreover, the bulk density resulted up to 30% lower and the energy behavior was improved up to 40%, making the developed mortars highly suitable for promising energy-saving uses. Finally, the waste recycling about halves the materials cost and could also grant further financial saving for the fish industry. To conclude, the large amount of reused bio-waste not only represents a valid alternative to their usual disposal in landfills, but also makes the considered mortars suitable for building applications and promising candidates for the Minimum Environmental Criteria certification, in light of the EU Green Transition, and in line with the principles of the circular economy.

Keywords: bio-composite mortar; construction; mollusk shell; waste recycling; circular economy; sustainability

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1. Introduction

This study is based on the main consideration that nowadays construction, among the various industrial sectors, is recognized worldwide for its major unsustainability, caused by the massive use of non-renewable raw materials (i.e., sand, soil, clay, minerals, etc.) and energy consumption. At the same time, this sector generates amazing amounts of green-house gases and wastes [1–3]. However, building activities are estimated to rise in the next decades and the need for new structures and infrastructures is associated with the increasing manufacturing of low-cost and high-performance materials [4]. In this regard, ordinary Portland cement (OPC) is one of the most produced construction materials globally because of its extremely low cost, easy manufacturing, and effective performance [5]. Nevertheless, cement production is extremely polluting (i.e., the manufacture of one ton of cement releases about one ton of CO_2 in the atmosphere), as is its associated industry (i.e., aggregate preparation) that strongly contributes to the general unsustainability of the sector [6,7].

In the last decade, many regulations have been issued to limit such problems i.e., the recent European "Renovation Wave", a flagship initiative under the EU "Green Deal" and the "Blu BioEconomy", particularly dedicated to the construction sector, the United Nations call for action "Sustainable Development Goals" (SDG), etc. Hence, developing greener and more high-performance materials, improving the use of secondary resources, fostering waste incorporation, and—even more—implementing sustainable cost-effective and greener manufacturing processes are highly essential.

At the same time, the rapid growth of the world population is leading to an exponential increase in the demand not only for new houses and infrastructures but also for food and energy resources, accelerating the growth of the agri-food industries. On the other hand, such massive production generates, once again, huge quantities of food waste—estimated to reach 2.6 million tons by 2025 globally [8]—whose treatment and disposal is often complicated and shows enormous environmental footprints, apart from the potential risk for the environment and humans caused by the decomposition of the organic matter [9]. Hence, a wiser and more careful waste management can lead to a considerable reduction in the environmental impact, a significant financial saving, and—moreover—an improved circular economy (CE) [10,11], thus providing a valid alternative to the usual disposal in landfill [12,13].

Having that in mind, this experimental study will try to answer to the demand of novel building and construction materials by reusing organic wastes. More particularly, this paper analyzes the possibility of recycling mussel shells (MS), deriving from a local fish canning industry, as a unique aggregate in total substitution of sand, which in this way can be potentially preserved in the manufacturing of greener OPC-based mortars in light of the CE approach. Moreover, the suggested massive reuse of agri-food wastes will be proposed as a valid alternative to their usual disposal in landfill.

Among the food industries, mollusk production is continuously increasing worldwide with China, Chile, and Spain leading the market [14]. Mollusk waste generally derives from three sectors: fishing and breeding, food processing, and trade and consumption. The mollusk production process takes place through a series of phases from mollusk breeding, to the harvesting and treatment, and packaging and marketing of the final food products. In 2019, the world's mussel production reached about 2 million tons, while in Europe it represented over two-thirds of total aquaculture production, reaching 487,662 tons with a value of about EUR 451 million [15]. However, such massive production corresponds to a consequentially large waste amount, mainly in the form of shells, which can represent up to 90% of the live mollusk's weight. Shell waste is a major problem for shellfish producers, sellers, and consumers, both in practice and economically. Shell disposal procedures are frequently unregulated and can cause many environmental concerns in terms of unwanted odor and contamination of the area, caused by the decomposition of the organic matter along with visual pollution [16]. In Europe, mollusk shells are considered animal by-products (ABPs) whose management and treatment are governed by the European Regulation 1069/2009/CE [17] in order to avoid any risk to public health and animal health, as well as to protect the safety of the food and feed chain [18].

The scientific literature reports the reuse of the various agri-food wastes for the production of novel building and construction materials. However, the scientific interest in using mollusk shell recycling in novel materials manufacturing is very recent and little has been written about the development of building and construction materials. Nevertheless, bivalve mollusk shells are mainly constituted of calcium carbonate ($CaCO_3$), the main component of many building materials, and have the potential to become a promising secondary raw material for several applications in construction. Clam shells were studied as coarse aggregate in green concrete production [19], oyster shells were reused as aggregate in partial substitution to sand [20] or in lightweight red bricks manufacture [21], fan-shaped shells, on the other side, were finely crushed and used as filler [22]. MS found some applications as aggregate, in partial substitution to sand (25%, 50%, and 75%) in aerial lime mortars [23,24].

These investigations, along with the presented one, clearly show the potential of reusing shell wastes in the construction industry to encourage the construction of green buildings, mainly offering economic benefits and improved environmental prospects. However, still today, hardly any kind of mollusk shell is completely recycled to produce and market green construction materials because it is generally considered an ABP waste and, consequently, landfilled or discarded into the sea. Only a few building and design products are present on the market, such as natural stones and tiles for landscape architecture [25], external paving [26], and even crockery [27].

Hardly any studies were found about MS recycling in the construction of OPC-based materials, moreover, with a reasoned technological characterization aimed at their possible real reuse in construction. Hence, the present work is highly innovative as it considers a whole replacement of the unrenewable traditional aggregate (sand) with MS. Moreover, a complete engineering characterization is performed analyzing how various MS granulometries could be used to design different mixes and understand the implication of the manufacturing procedure on the final materials' performance. Finally, this is the first time that the cost of MS-based materials is predicted both for the hypothetical manufacturing company and the fish canning industry, considering the possible financial profits of the two.

All considering, this experimental work represents a substantial advancement of the state of the art, focusing on a highly common building and construction material (cementitious mortar) that needs a strong boost of sustainability, and shows a character of high innovation for a greener and more efficient building sector, in light of the CE approach and according to the recent EU directives.

2. Material and Methods

2.1. Materials

In this study, the novel green bio-composite mortars were developed by using a conventional construction binder, and MS as aggregate in complete substitution to the traditional commercial sand. The used raw materials were:

- OPC, branded i.work Tecnocem 32.5 R (CEM II/B-LL) by Italcementi (CE marked in compliance with UNI EN 197-1:2011 [28]) was used as binder;
- natural siliceous sand by Axton (CE marked in compliance with UNI EN 13139:2003 [29]) with a standard calibrated granulometry of 0–4 mm, was used as benchmark fine aggregate;
- MS, collected as waste deriving from a local fish canning industry, was used as fine aggregate in substitution to sand;
- drinkable water taken from the municipal aqueduct was used in compliance with UNI EN 1008:2003 [30];
- releasing agent CS Distak by Gattocel Italia (a natural resin emulsion) was used to facilitate the extraction of the specimens from the molds, preventing their adhesion.

2.2. Product Mix Design

The bio-composite mortars' mix design was prepared according to the standard EN 998–2:2016 as follows: 1 part of binder, 3 parts of aggregate, and 1 part of water (volumetric parts, V.%). MS were used in various granulometric ranges in compliance with EN 13139:2013 [29]. In this paper, an ordinary OPC-based mortar was prepared for comparative reasons as reference material by using a commercial natural siliceous sand (standard granulometry of 0–4 mm); hence, an analogous MS granulometric calibrated fraction was prepared to simulate the same composition of the used commercial sand. A complete description of the aggregate composition of each mortar mix is shown in Table 1. The table also reports both the water (W) to binder (B) and the liquid (L) to solid (S) ratios.

Table 1. Experimental plan of the produced bio-mortars.

Specimen n.	Aggregate Nature	Granulometric Range [mm]	W/B [wt.%]	L/S [wt.%]
1	Ref. (sand)	0.00–4.00	0.714	0.149
2	MS	0.00–4.00		0.178
3	MS	0.00–1.00		0.183
4	MS	1.00–2.00		0.192
5	MS	2.00–4.00		0.201

Finally, each mix was tested to assess the influence of the wastes' granulometry to the mortars' engineering features, investigating the possibility of producing a uniform material, and evaluating the possible uses in construction, according to the relevant standards and guidelines (i.e., EN 998-2, Eurocode 2, Italian D.M. 17/01/2018, ACI 116R-00).

2.3. Processing Details

Before manufacturing the mortar mixes, some pre-treatments were implemented on the waste-based aggregate. The supplied MS contained some organic matter and salt residues on their surface. Consequently, before being used, they were subjected to the following pre-treatments:

(A) Cleaning: furnished MS (Figure 1A) were previously cleaned manually with brushes and sponges to remove any organic residues; subsequently the MS were washed with running water to remove any traces of dirt and/or remains, as well as the possible sea salt residues (Figure 1B,C).

(B) Drying: the MS were dried in a conventional oven at a temperature of 60 °C for 24 h to remove any content of water, to obtain a completely dry product, and to facilitate the following step (Figure 1D).

(C) Grinding: the dried MS were manually ground by hammer to roughly crush the shell and reduce them into smaller pieces, then the grinding process was continued by hand by thinly grinding the small shell pieces in a ceramic mortar to obtain a fine aggregate with a granulometry $\leq$ 4.00 mm (Figure 1E).

(D) Sieving: the sieving phase was carried out using standard metallic sieves (UNI EN 13139:2013 [29]) (Figure 1F) to obtain the desired selected granulometries (0–1 mm, 1–2 mm, 2–4 mm).

(E) MS selected granulometries were mixed to obtain a similar granulometric calibrated range 0–4 mm of the commercial sand.

(F) Packing: the prepared MS granulometries were hermetically packed in a plastic bag.

The packed commercial sand presented a 12 wt.% moisture content, therefore, before being used, it was dried in a conventional oven for 24 h at a temperature of 60 °C to avoid compromising the designed water/binder ratio. It was then naturally cooled down and hermetically packed in a plastic bag before use.

In this work, mortars were manufactured following the standardized procedure of UNI EN 196-1:2016 [31], and at ambient conditions (20 ± 2 °C, 65% RH). The process consisted of simple, safe, reproducible, and sustainable steps.

Mixes were prepared using a programmable automatic mixer (Automix mod. 65-L0006/AM by Controls). Raw materials were poured into the mixer bowl in the following sequence (Figure 2A): (1) water, (2) binder, and lastly (3) aggregate. In the first phase, while pouring the aforementioned raw materials, the mixer rotated at 140 rpm; subsequently, after 1 min, speed was increased to 285 rpm for 30 s immediately followed by a 90 s pause to allow the water to be absorbed by the binder particles. Afterwards, the mixer was started again at 285 rpm for another 60 s up to completion.

Figure 1. Pre-treatments on the MS: (**A**) furnished shells, (**B**) surface cleaning, (**C**) washing under running water, (**D**) drying, (**E**) grinding, and (**F**) sieving.

The fresh material (Figure 2B), in accordance with UNI EN 1015-11:2019 [32], was poured into standard metallic molds (40 × 40 × 160 mm), that had been previously brushed with the releasing agent (Figure 2C). The slurry was properly set by 25 pestle strokes, followed by slight vibrations. This operation, beyond uniformly distributing the slurry itself through the available space, also prevented the formation of voids in the specimens that could alter the testing. At the end, the leftover mortar was removed with the aid of a metallic spatula, in order to obtain a flat upper surface, and levelled to the mold edge.

Once the slurry was carefully poured and set into the standard molds, specimens were cured, in compliance with the same UNI EN 1015-11:2019 [32], at ambient temperature by following the next consecutive phases:

(1) days 1–2: during the first 2 days, the molds—filled with the fluid slurry—were kept closed in plastic bags to grant 95 ± 5% RH (Figure 2D);
(2) days 3–7: once the slurry was hardened, the specimens were taken out from the molds (if some specimens were still wet/viscous, they were kept in the molds for another 24 h), put again in the same bags, and kept closed for the following 5 days to grant the same curing conditions (Figure 2E);
(3) day 8: the specimens are extracted from the bags and stored at ambient conditions;
(4) day 8–28: specimens were cured at ambient conditions until testing (Figure 1F).

Figure 2. Mortars manufacture: (**A**) mixing, (**B**) spatulating, (**C**) pouring in standard molds, (**D**,**E**) curing, and (**F**) unsealing.

2.4. Materials Characterisation

The wastes' (MS) mineralogical composition was determined by X-ray powder diffraction (XRD) using a Rigaku Geigerflex D/max-Series instrument, Tokyo, Japan (Cu Kα radiation, 5–80° 2θ range, 0.026° 2θ step-scan, and 2 s per step). For this analysis, the samples were ground to a particle size inferior to 63 μm. The chemical composition was evaluated by X-ray fluorescence (XRF) using a Phillips X'Pert PROMPDspectrometer, Almelo, The Netherlands. The loss on ignition (LOI) (at 1000 °C for 15 min) of the dried samples was also measured. Bulk density was calculated from the Archimedes' principle, as the average of three tests. Optical images were taken by a microscope using the Nikon mod. P-SXY64, Tokyo, Japan. The microstructure was investigated by scanning electron microscopy (SEM) using a Hitachi analytical FE-SEM SU-70.

The consistency of the fresh slurry was estimated by flow table test in accordance with EN 1015-3:1999 [33], measuring the fresh slurry spread on an automatic flow table, model 63-L0037/E by Controls. Bulk density was calculated at 28 days of curing from geometric calculations, as the average of three specimens. The water imbibition (weight variation, ΔP/P %) and the capillary index were determined according to EN 1015-18:2004 [34]; the specimens were formerly dried to constant mass in a conventional oven at 60 °C. The given values are the average of three measurements for each formulation. The mechanical performance was determined according to UNI EN 1015-11:2019 [32] and UNI EN 998-2:2016 [35] using a universal testing machine (Controls mod. Automax 5) equipped with a 250 kN load cell. The bending resistance (BR) was calculated by a 3-point bending test with the cell running at a rate of 0.50 MPa/s, and with a support span of 100 mm; the uniaxial compressive strength (UCS) was evaluated with the cell running at a rate of 0.75 MPa/s. The given values are the average from each of the three tests. The thermal conductivity (λ) was measured using an HFM-CT 1000 calorimeter in accordance with the UNI EN 12667:2002 [36]. Three tests were carried out for each formulation, and specimens were previously dried at 60 °C for 24 h in a conventional oven to eliminate the internal moisture.

3. Results and Discussion

3.1. Waste Characterisation

The XRD pattern of MS (Figure 3) shows that calcite and aragonite, two different $CaCO_3$ polymorph structures, are the main crystalline detected phases in line with [37–39]. Accordingly, CaO is the main constituent that was identified by XRF (cf. Table 2), and the high LOI value (44.91 wt.%) is due to the $CaCO_3$ thermal decomposition.

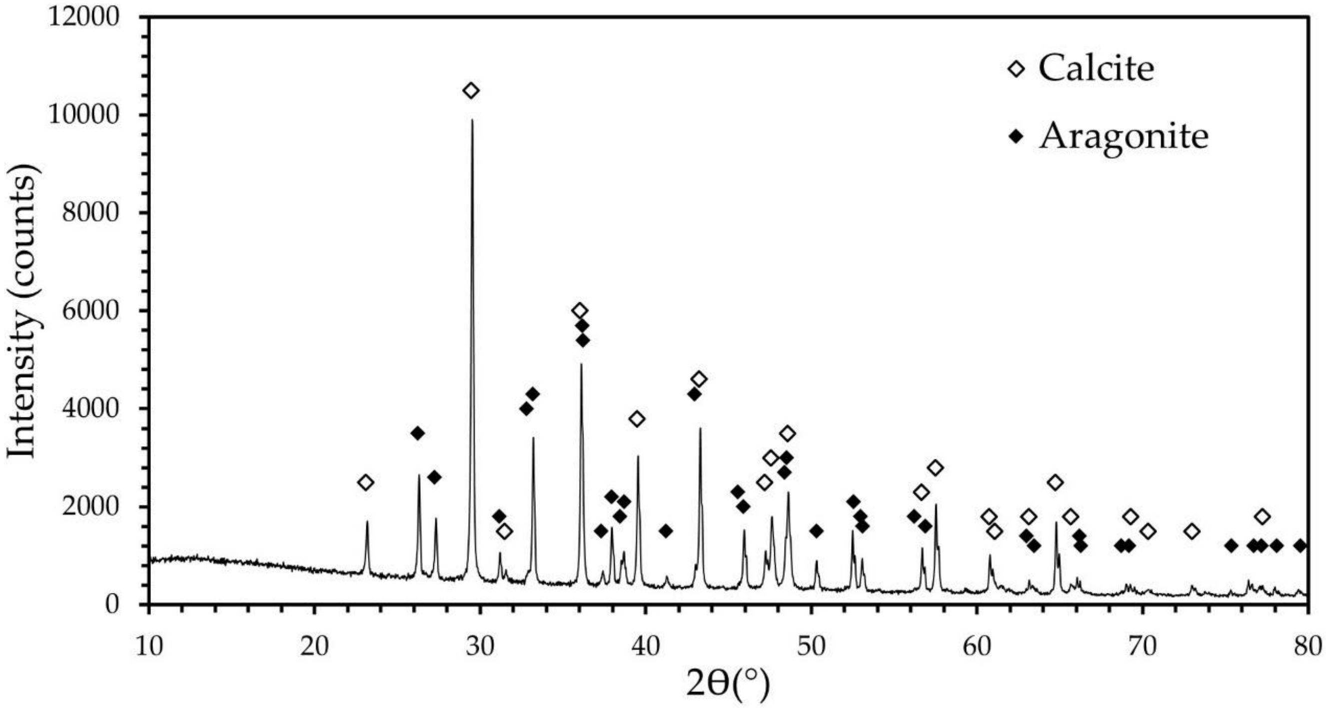

Figure 3. XRD patterns of MS.

Table 2. MS chemical composition, as estimated by XRF.

Oxides	[wt.%]	Oxides	[wt.%]
CaO	52.21	MgO	0.30
SiO_2	0.78	P_2O_5	0.09
Na_2O	0.74	Al_2O_3	0.09
SO_3	0.37	Sr	0.08
Cl	0.33	Fe_2O_3	0.06
LOI (1000 °C): 44.91 wt.%			

The bulk density of the MS—in the various granulometries—is shown in Figure 4, the optical images in Figure 5. It is observed that in increasing the particles' granulometry, the density diminishes. That is due to the larger spaces between the shell fragments. In comparison, the calibrated 0–4 mm MS aggregate presents a higher density as, reasonably, the smaller fractions (0–1 mm) tend to occupy all the available voids. In any case, the latter results were much lighter than commercial sand. Finally, the whole shell's density was also measured, resulting in the highest value (even though the entire shell cannot be used due to its large dimensions that are not suitable for architectural applications).

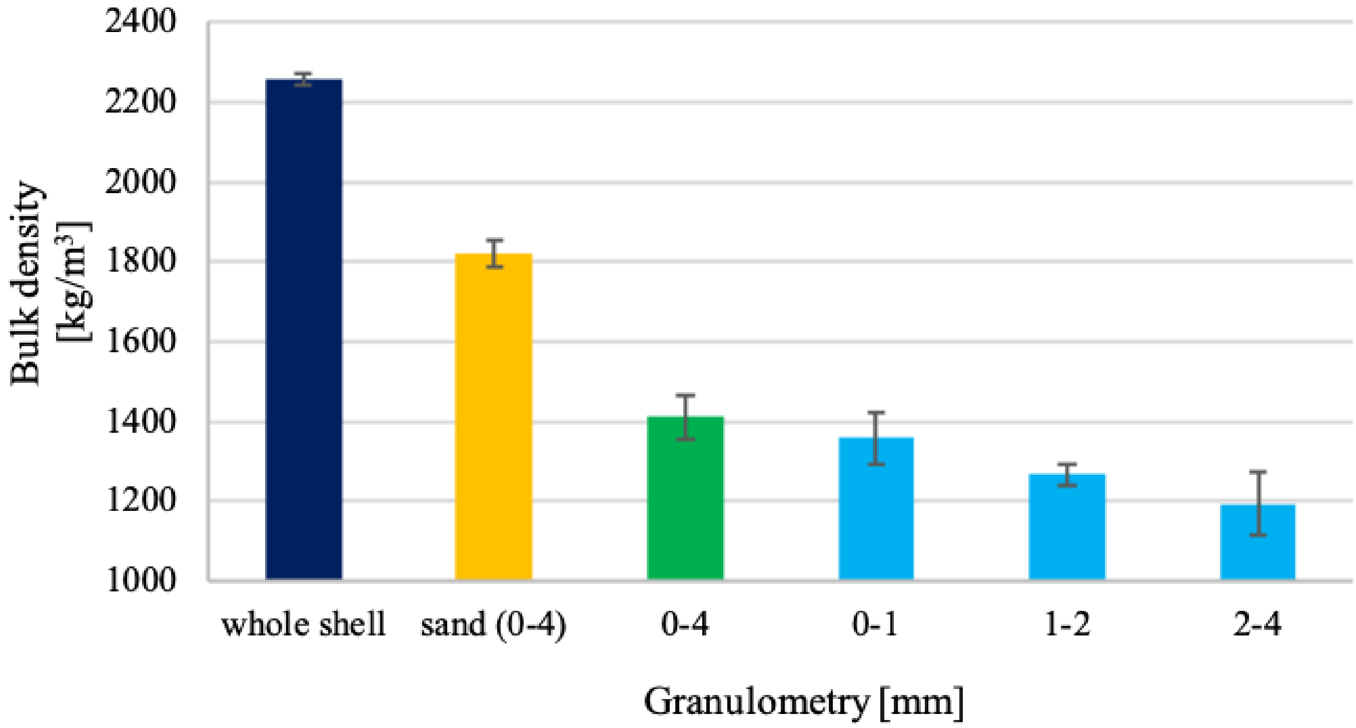

Figure 4. MS bulk density.

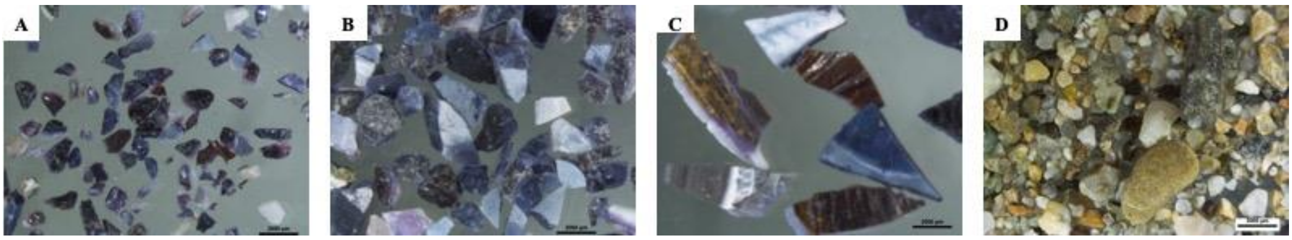

Figure 5. Optical imaging of the calibrated MS ((**A**)—0–1 mm, (**B**)—1–2 mm, (**C**)—2–4 mm), and the commercial sand (**D**).

Optical images reveal the morphology of the selected MS fragments (Figure 5A–C). They present an irregular flat shape, with sharp edges and pointed corners. Smaller fragments appear smoother and round. For comparative reasons, an optical microscopy image of the commercial sand is shown (Figure 5D) that, conversely, presents a pseudo-spherical shape with rounded and smooth edges. Figure 6A shows the MS morphology of the surface that, in accordance with the previous observations, appears extremely flat and compact with tiny rough spots. In Figure 6B,C the interphase MS-mortar and the reference mortar matrix are shown, respectively. As can be seen, due to the MS morphology, the contact surface between the binder and the shells (waste aggregate), although homogeneous and well distributed, is nevertheless neat and extremely well defined, without intersections and interconnections between the granules. That, as it will be discussed next, limits the friction between the two phases generating some issues for applicability, especially in the materials' mechanical resistance. The results of the mortar matrix (Figure 6C), as expected, were homogeneous and well assorted, with the sand particles well distributed and completely covered by the binder.

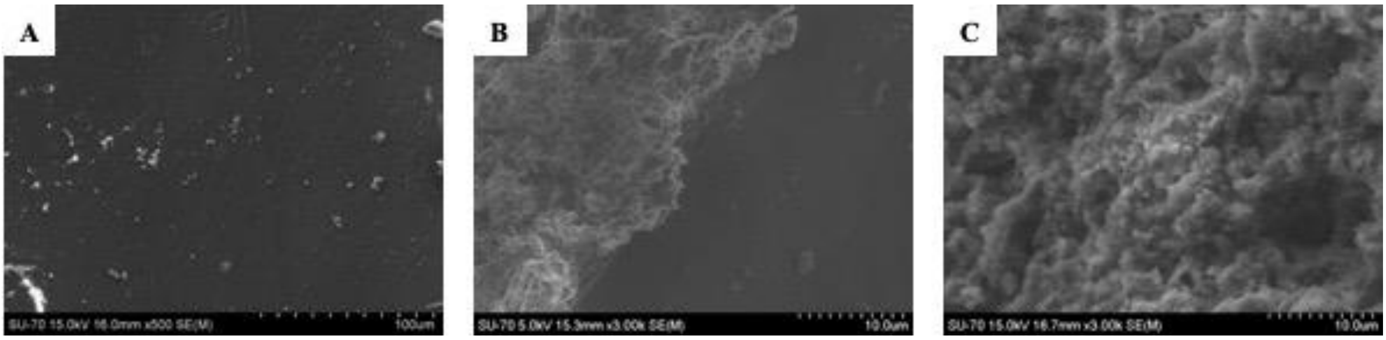

Figure 6. SEM imaging of: (**A**)—MS surface (×500), (**B**)—interface MS-mortar (×3000), and (**C**)—reference mortar (×3000).

3.2. Mortars Characterisation

The final products' characteristics strictly depend on the designed mix, on the raw materials nature, and on the followed manufacturing process. All of that deeply contributes to the achievement of a material with suitable features for the desired applications, in our case architectural and constructive ones.

Having assessed the raw materials features, especially when dealing with waste-based resources, an improper mortar mix design may cause many issues in setting, installation, or even a complete failure. On the contrary, an accurate mix is crucial to achieve the required blending, the strongest bond possible, the saturation of the individual molecules, maximum uniformity of the components, and thus, a long-lasting installation, set, and cure.

In all the following graphs (Figures 7–11) the reference cementitious mortar (sample n. 1) is colored in yellow; the calibrated 0–4 mm MS-based mortars (sample n. 2) in green; the selected 0–1 mm, 1–2 mm; and 2–4 mm MS-based mortars (samples n. 3–5) in light blue.

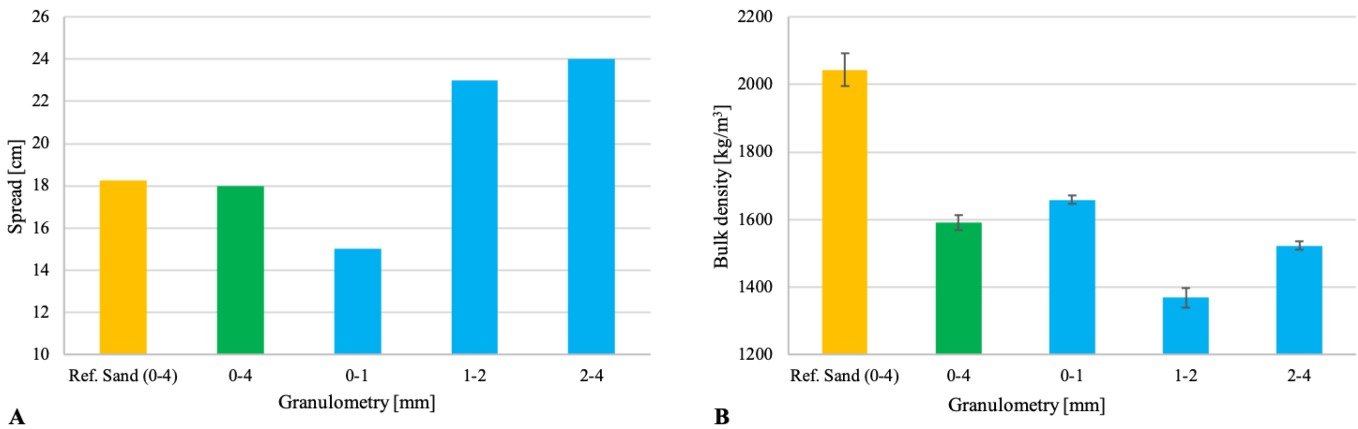

Figure 7. Measured spread (**A**); bulk density (**B**).

Macroscale Properties

Figure 7A shows the workability of the produced mortars and Figure 7B shows the mortars' bulk density. The slurry consistency is fundamental for predicting the possible applicability of the material, indicating its attitude to be uniformly mixed and placed to avoid any engineering issues (i.e., presence of voids, segregation, impossibility/delay to set properly [40,41]. In this study, consistency was evaluated by measuring the slurry spread on a flow table (flow table test). A good workability for applications in construction is, generally, assured by an 18–22 cm spread, still suitable for border values. Extreme values (10–15 cm and 25–30 cm) indicate a slurry that is too pasty/fluid, respectively, showing an inadequate consistency. Usually, consistency can be controlled by adding chemicals (i.e., plasticizer), however, in this study they were not used to isolate the influence of the MS granulometry on the final product.

As expected, the mortars' workability was quite reduced by using very thin aggregate (MS 0–1 mm) as the small MS particles tend to occupy the paste voids, making it pastier. Moreover, the smaller particles need more water to lubricate their surface, which diminishes the mixture's workability. On the other hand, by increasing the MS granulometry, the workability increases. In fact, the opposite behavior was verified for the 1–2 mm and 2–4 mm granulometries, as expected, which increased the mixture's spread. The flatness and irregular shape of these particles compromise the compactness of the slurries in the mold, despite the better spread (that results from more available kneading water), decreasing their density and consequently resulting in higher water absorption. The calibrated MS 0–4 mm shows values much smaller than the selected MS granulometries (1–2 mm and 2–4 mm), while they are comparable to those of the commercial sand, indicating that the workability of a traditional mortar is similar to the considered waste-based one. That was partially expected, as the aggregate granulometric range is one of the major factors affecting such property; here, the MS calibrated 0–4 mm granulometric range was prepared to simulate the sand's one, hence there is a direct relation between the two.

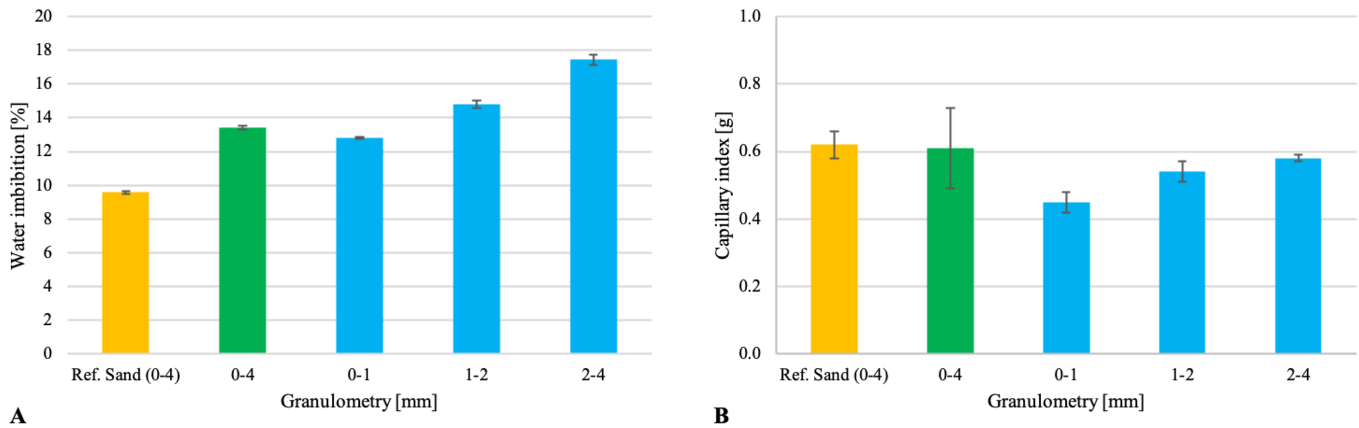

Figure 8. Water sorptivity: water absorption by imbibition (**A**); capillary index (**B**).

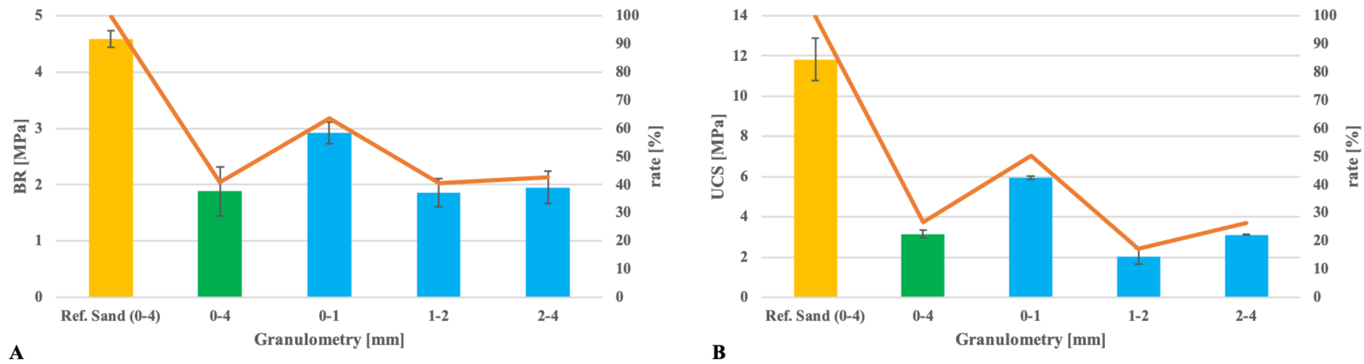

Figure 9. Mechanical performance of the mortars: bending resistance (**A**); uniaxial compressive strength (**B**).

Bulk density varies a lot depending on the used MS aggregate. All of the specimens' results were much less dense than the reference mortar, in accordance with the aggregate density that is lighter than the sand (cf. Figure 4). Among the MS-based mortars, specimens containing the smallest MS fraction (0–1-mm) results were denser, showing a higher mass as the fractions were designed in a ration based on volume %.

The behavior toward water is shown in Figure 8A (imbibition) and Figure 8B (capillary action). It can be observed that all the MS-based mortars present higher values of water imbibition in comparison to the reference one. Moreover, by increasing the MS particle size, the absorbed water increases. In particular, the calculated reference mortar's water absorption was 9.57%, while the calibrated MS-based mortar was 13.42%. That is explainable as the latter is less dense than the reference material (cf. Figure 7B), hence the specimen results more porous, causing more water to be absorbed and retained inside. Moreover, as no intersections were seen between the MS and the mortar matrix (cf. Figure 6B), it is expectable that among them some little space could be generated, thus permitting water to rise in. Such observed behavior is in accordance with other studies found in the literature [42] where the use of seashells as aggregate in mortar manufacturing leads to an increase of the water sorptivity. Once again, this phenomenon has been related to the shells' porosity and mobility into the matrix (low friction due to smooth and flat surfaces, cf. Figure 6) which, in turn, generates an interconnected porosity in the material, as already hypothesized in this study.

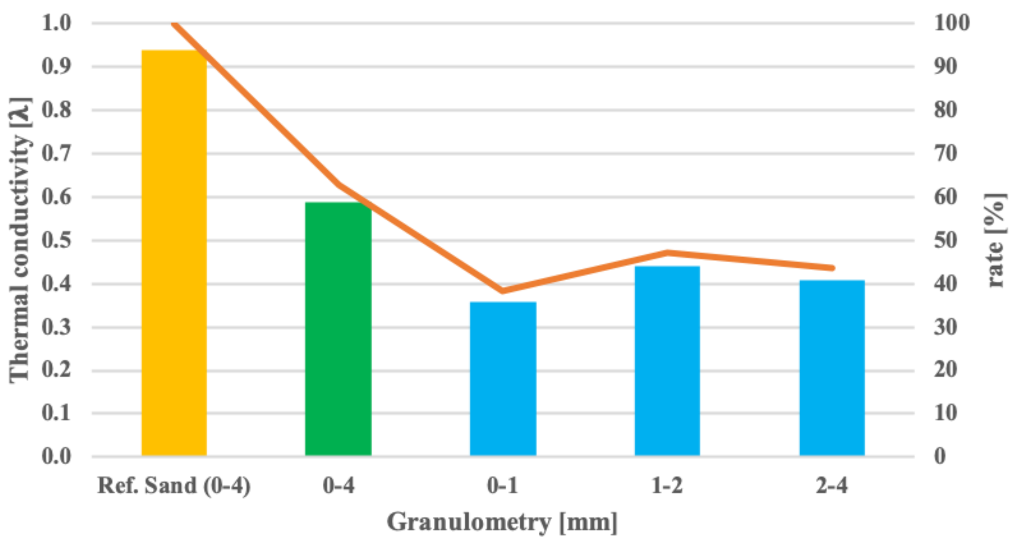

Figure 10. Thermal conductivity (λ) of the produced mortars.

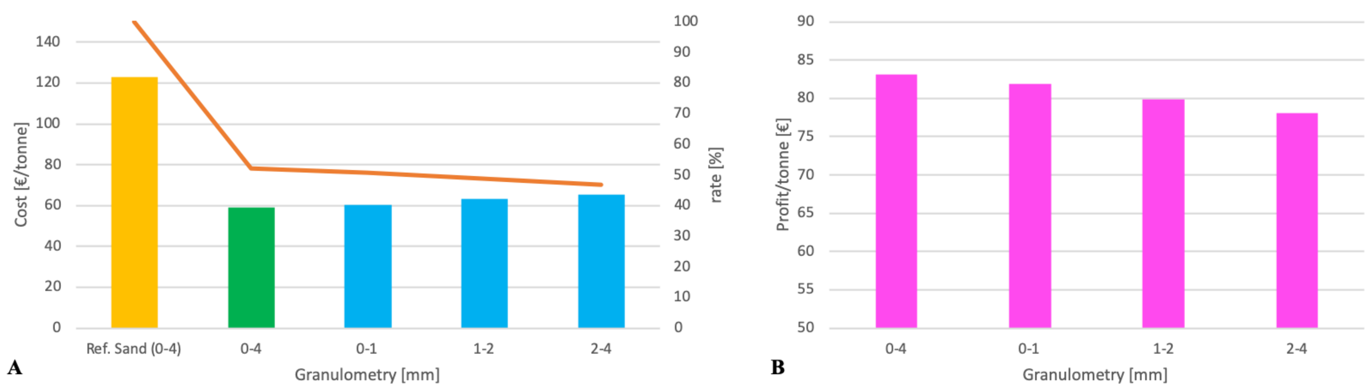

Figure 11. Material cost estimation for the manufacturing company (**A**); fish canning industry profit per furnished MS ton (**B**).

The capillary action, on the other hand, shows a more interesting behavior; the capillary indexes for the reference material and the calibrated 0–4 mm MS-based mortar are approximately the same: 0.62 ± 0.04% vs. 0.61 ± 0.12%, respectively. That is in contrast with the results reported in [43] where a partial substitution of sand with MS led to a substantial capillary uptake decrease, with the main difference being that in this novel study the sand vs. MS substitution was complete. That is easily explainable by the fact that in the aggregate blend of [43], the thinnest MS particles generate a more homogeneous mortars matrix, while a whole MS aggregate, as the one considered in this study, basically behaves as a traditional sand aggregate. On the other hand, selected MS granulometries lead to smaller capillary actions. In particular, the 0–1 mm MS-based mortar shows the lowest value because the very small aggregate particles tend to occupy the micro-voids within the mortar's matrix, preventing the water rise by capillarity. Interestingly, all the produced specimen results are suitable for applications in construction, because the water sorptivity is in line with the considered applications.

The mechanical performance of the specimens was assessed at 28 days of curing, and is presented in Figure 9A,B to show the bending resistance (BR) and the uniaxial compressive strength (UCS), respectively, along with the decrease rates compared to the reference mortar. It is observed that all the MS-based mortars show much less resistance, both BR and UCS, in comparison to the standard reference mortar. In particular, for all the MS-based mortars, the observed BR reduction results were rather homogeneous, with BR values equal to about 40% of the reference mortar. For the thinner granulometric range, the selected 0–1 mm MS-based mix (sample n. 3), the decrease rate is higher with results showing about 63% less than the reference material. The same trend is observed for the UCS values, but with generally lower decrease rates whose values were equal to about 25% and 50% compared to the reference material. That is probably caused by the MS' irregular, flat, and sharp shape that cannot assure a proper friction within the binder paste, making the mortar weaker. That is also in accordance with the optical microscopy and SEM (cf. Figures 5 and 6), and at the same time, they are also less dense (Figure 7B) and more porous (Figure 8A).

The obtained results are also in line with the studies of Martínez-García [23,24] where MS were reused as fine and coarse aggregate in aerial lime mortar manufacturing for coating applications. There, it was observed that the MS' irregular particles morphology generated large pores in the mortar's matrix, beyond being less dense and porous, as also seen in the present study. As a direct consequence, the bond among the MS particles and the binder was highly reduced, compromising the overall mechanical strength. Accordingly, other authors have already observed that, in general, seashell-based aggregates could decrease materials' mechanical performance due to the shells' smooth surface area [42,44]. To conclude, our study confirms the general thought that reusing mollusk shells could reducing mortars' mechanical strength, which could be a real concern, especially when developing novel construction materials.

UCS values of all the formulations are reported in Table 3, along with the relative classes of resistance in accordance with UNI EN 998-1 [45] (plastering application) and UNI EN 998-2 [35] (masonry application). It is noted that none of the MS-based mixes are suitable as structural mortars for concrete applications (class M > 10), in accordance with the above discussion, while the results are adequate for light structures or bed joints, with the exception of mix n. 4 (1–2 mm) whose UCS is too low to be considered for any of such applications. That consideration was highly interesting, as the investigation conducted so far would allow us to predict some light structural applications for mollusk shell reuse in construction rather than discarding them a priori. On the other hand, all the produced MS-based mixes were highly suitable for plastering/coating applications, as underlined by previous studies [23,24].

The produced materials' energy-saving performance was quantified measuring the thermal conductivity (λ) that returns the heat flow through the material. The measured λ values are presented in Figure 10. It is observed that all the developed MS-based mortars present a lower λ in comparison to the reference mortar. In particular, among the waste-based materials, the calibrated MS 0–4 mm mix showed the higher value, with a reduction of about 40% to the reference material. The lowest λ value was measured for the selected MS 0–1 mm, with a nearly 60% λ reduction. That is explainable by the porous and light nature of the mollusk shells that, in contrast to the more compact and dense sand, acts as a sort of light aggregate, hence with a more insulating performance that is transferred to the developed mortar.

Table 3. UCS values for each mortar and relative class of resistance.

Specimen n.	Aggregate Nature	UCS [MPa]	EN 998-2 [35] (Masonry)	EN 998-1 [45] (Plaster)
1	Ref. (sand 0–4 mm)	11.83	M10	CSIV
2	MS (0–4 mm)	3.15	M2.5	CSII
3	MS (0–1 mm)	5.95	M5	CSIII
4	MS (1–2 mm)	2.04	M1	CSI
5	MS (2–4 mm)	3.11	M2.5	CSII

(NB. Standard deviation values were negligible and are not reported in the table).

In any case, it can be concluded that all the considered materials perform much better than the traditional cementitious material. In other words, if they were applied in a real building (i.e., as large surfaces' plastering) they would grant a higher thermal insulation and, consequently, much more financial savings in the building energy management. That is also in line with [46], where energy simulations on a building model revealed a 28.3% financial savings in comparison to standard materials. That stresses the necessity to further study, produce, and apply such novel materials with promising energy applications. In addition to these financial savings, the energy savings associated with the enhanced insulating properties also have to be considered (see Figure 10). Therefore, it can be stated that MS valorization and reuse could become a powerful resource in building construction, especially to improve energy management.

4. Materials Cost Estimation

In this section, a preliminary estimate of the materials' cost is performed on the basis of the raw materials' quantities for each developed mix, considering a possible mortar manufacture of 1 ton. Prices for the raw materials, actualized to the current year from a market survey, are the following: OPC—250,00 EUR/ton; H_2O—40,00 EUR/ton; sand—105,00 EUR/ton; SCG—250,00 EUR/ton. The costs associated with MS disposal include transport and landfill, while costs associated with MS recycling include transport, laboratory analysis, and preparation (washing, grinding, and sieving). Transportation costs were taken from the Italian Ministry of Infrastructure and Transportation consider trucks of category C (>12 tons) that appear to be the most appropriate in pondering a potential mass production. The other costs derive from market surveys.

It is noted that both the manufacturing company and the fish canning industry could gain financial advantages from waste recycling. In fact, the former could obtain the MS residue at no cost, while its transportation and aggregate preparation (washing, crushing, and sieving) remains at its own expense. Therefore, to calculate the final materials costs, waste treatment costs are summed up to the initial expenditure (it is noted that no sand is used for the waste-based mixes). The latter generates profit by avoiding the residues in landfills that, conversely, is to be considered as the financial benefit instead of a fixed outflow. The results of these calculations are shown in Table 4—for the reference material and the calibrated MS 0–4 mm mix; Figure 11A,B shows the materials costs/benefit for the two industries and the final product price and EUR% reduction. Note that the calculations are based on the actual MS quantities needed for the various mixes (depending onto the MS-aggregate density).

Table 4. Raw materials quantities [wt.%] involved in the process, relative cost [EUR/t] and indication of the final product price/profit.

Reference Mortar—Sand (0–4 mm)							
Company	**Activity**	**Description**	**Unitary Price**	**U.M.**	**Fraction**	**Partial Cost [€/t]**	**Total Cost [€/t]**
Manufactury	**raw materials**	OPC *	250,00	€/t	0.18	45,40	
		H2O *	40,00	€/t	0.13	5,19	
		Sand *	105,00	€/t	0.69	72,32	
	MS recycling	MS-transport/km **	0,12	€/t × km	0.00	0,00	
		MS-chemical analysis ***	50,00	unitary	0.00	0,00	
		MS-preparation ***	5,00	€/t	0.00	0,00	**122,90**
Fish canning industry	**MS disposal**	MS-transport (1 km) **	0,30	€/t × km	0.00	0,00	
		MS-landfill ***	130,00	€/t	0.00	0,00	**0,00**
NB: * market price; ** Italian Minitry of Infrastructure and Transportation; *** market survey							
Calibrated 0–4 mm MS-Based Mortar							
Company	**Activity**	**Description**	**Unitary Price**	**U.M.**	**Fraction**	**Partial Cost [€/t]**	**Total Cost [€/t]**
Manufactury	**raw materials**	OPC *	250,00	€/t	0.21	52,77	
		H_2O *	40,00	€/t	0.15	6,03	
		Sand *	105,00	€/t	0.00	0,00	
	MS recycling	MS-transport/km **	0,12	€/t × km	0.64	0,08	
		MS-chemical analysis ***	4,00	unitary/cargo	0.64	2,55	
		MS-preparation ***	5,00	€/t	0.64	3,19	**64,62**
Fish canning industry	**MS disposal**	MS-transport (1 km) **	0,30	€/t × km	0.64	0,19	
		MS-landfill ***	130,00	€/t	0.64	82,96	**83,15**
NB: * market price; ** Italian Ministry of Infrastructure and Transportation; *** market survey							

Relating to the mortar manufacturing industry, it can be observed that for all the mixes, the complete substitution of sand with MS led to a financial savings of between 46.80% and 52%. In addition to these financial savings, the energy savings in building energy management, associated with the enhanced insulating properties, also have to be considered (see Section 3). On the other side, the fish canning industry can gain savings of between 78 EUR/ton and 83 EUR/ton by MS furnishing, thus avoiding any activity of recycling rather than simply discarding the waste. Therefore, it can be stated that MS could become a powerful resource in building construction and associate companies to improve energy management and reduce the costs associated with construction and waste management.

5. Conclusions

In this preliminary study, the possibility of manufacturing novel sustainable bio-composite mortars by reusing ground mussel shells in total substitution of sand is reported. The waste-based aggregate was tested in various granulometries: selected granulometric intervals 0–1 mm, 1–2 mm, and 2–4 mm; and a calibrated mix 0–4 mm to simulate the same as that of a traditional commercial sand mixture.

Results showed that recycling mussel shells in mortar manufacturing is feasible, and it is possible to prepare a green material suitable for various applications in construction.

In particular, the developed mortars are more appropriate for plastering applications rather than for structural ones, even though such application is not excluded. Moreover, the reuse of mussel shells led to a significant improvement in the energy behavior and, consequently, a more sustainable building management. To this, high financial savings could be generated for both the manufacturing and the fish canning industries, making MS recycling also profitable from a company's financial point of view.

The novel bio-composites were prepared with the same simple protocol that is generally used for the traditional mortars, making them suitable for real applicability in a real construction site and commercial development.

Ultimately, the reuse of such a bio-waste in substitution to sand would mean saving a non-renewable raw material, while valorizing a waste that is generally landfilled with many environmental concerns in light of the circular economy approach and improved sustainability.

Further investigations will focus on the reuse of other types of mollusk shells with different binders to achieve a complete abacus of possibilities for application and development. An industrial scale-up will also be studied, with the aid of a life cycle assessment approach, to better design a proper industrial plant, improve the manufacturing procedure (especially in the waste pre-treatments), and study the real environmental footprint. Finally, tests on durability will be performed in order to understand the possible technological limit to the proposed application.

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