

Review

Trends and Opportunities of Bivalve Shells' Waste Valorization in a Prospect of Circular Blue Bioeconomy

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Abstract: Bivalves aquaculture is already considered a very sustainable for of food production and might become an essential pillar on which to develop future global food security. However, with the increase in production, a correspondingly great amount of waste will be produced all around the earth, principally in the form of shells, which can represent up to 90% of the fresh mollusk weight. Nowadays, shell waste has no notable use and is commonly regarded as waste, often dumped in landfill, or thrown back into the sea, causing a significant level of environmental concern, and resulting in a loss of natural and valuable resources. Bivalve shells are mainly formed by CaCO₃, giving them the potential to become a promising secondary raw material for several applications, from a circular economy perspective. This paper aims to review the scientific literature of the last twenty years and the principal recent trends in shell waste reuse, describing applications that are already in place on a large scale in agriculture and livestock, soils, water and wastewater remediation, biodiesel production and biofilters, as well as niche applications that now simply have the potential to be scaled up.

Keywords: circular economy; bivalves; shell waste; reuse; waste valorization



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1. Introduction

The notion of an expected rise in global population from 7.9 billion in 2022 [1] to 9.7 billion in 2050 has received growing attention because of its impact on world food security [2]. Even though the rate of increase in world population has been slowing for the last 50 years, from 2.2 to 1.05% per year, the growth in food demand is increasing because of the increase in income per capita, which acts as a new driver of food demand, especially in developing countries [3]. Moreover, we are currently witnessing a progressive shift towards animal-based products, putting further pressure on agricultural resources and overall food sustainability [4,5]. The endurance and resilience of food systems at local and global levels is a primary goal, even though, at present, this goal is strongly jeopardized by the finiteness of resources and agricultural soils, overall food losses and food waste [6]. The importance of these challenges is so crucial that they are part of the Agenda 2030 Sustainable Development Goals; namely, Target 2.4 states: "by 2030, ensure sustainable food production systems and implement resilient agricultural practices that increase productivity and production, that help maintain ecosystems, that strengthen capacity for adaptation to climate change, extreme weather, drought, flooding and other disasters and that progressively improve land and soil quality" and Target 12.3: "by 2030, halve per capita global food waste at the retail and consumer levels and reduce food losses along production and supply chains, including post-harvest losses" [7].

Despite this, the FAO State of Agriculture report (2021) estimates that global food loss and waste still represent one-third of globally consumed food per year [8]. In fact, about 14% of global food production is lost during supply chain stages, including in retail, and

about 931 million tonnes (121 kg per capita) of food waste is globally produced, allocated to household consumption (61%), food service (26%) and retail (13%) [9]. About 8–10% of global greenhouse gases emissions are attributable to food that is not consumed [10].

In recent years, the term *blue economy* has become a concept related to the sustainable utilization of marine resources. The limitation of cropland is pushing to look beyond green economy and toward the seas and oceans as potential immense repositories of resources [11]. Therefore, world fishery and aquaculture has undergone a remarkable expansion and is projected to reach over 200 million tonnes by 2030. Globally, fish accounts for about 17% of animal protein intake, growing at an annual rate of 3.1% and overtaking all other animal proteins, which increased by an average of 2.1%. In per capita terms, fish consumption arose from 9.0 kg in 1961 to 20.5 in 2018 and is projected to reach 21.5% by 2030 [12].

In 2018, world aquaculture fish production reached 82.1 million tonnes, dominated by finfish (54.3 million tonnes, 8.1 kg per capita) and mollusks, mainly bivalves (17.7 million tonnes, 2.6 kg per capita). Of the latter, more than 70% is represented by oysters, clams, scallops and mussels [13]. Clams and oysters are the main species, providing 38% and 33% of the overall production, whereas scallops and mussels contribute to 17% and 13%, respectively [14].

In fisheries and aquaculture, it is estimated that 30–35% of the global harvest is either lost or wasted every year, principally at the consumption stage in high-income countries, whereas in low–middle income countries, mainly because of inadequate preservation infrastructure [15], losses can take place along the entire supply chain, from capture and harvest to post-production (i.e., landing, handling, storage) or distribution [16].

For mollusks, a large volume of pre- and post-consumer residues is generated, mainly shells, which usually account for about 65–90% of live weight depending on the species. For the sole bivalves, over 10 million tonnes of shells are produced each year and generally dumped either in open fields or in landfills [17]. Even though mollusk cultures can provide several ecosystem benefits, i.e., anthropogenic CO₂ control, natural coastal protection, biodiversity maintenance, the principal barrier to its sustainable development is precisely the question of shells' waste management: at harvesting, depuration and post-consumption levels [18].

Shell waste is a major issue for shellfish producers, sellers, and consumers, both in practice and economically. Shell piles are frequent around the world due to unregulated disposal procedures, causing environmental harm in terms of an unwanted smell and area contamination caused by organic matter decomposition and visual pollution [19]. Since mollusk aquaculture is cited as low-impact food source, all aspects of production should be taken into consideration without overlooking the fate and management of shell waste. Improving the sustainability of the aquaculture sector through the implementation of waste reduction and/or valorization strategies will contribute to ensure food security over the coming decades [18].

Not surprisingly, food waste is one of the targeted areas in the Circular Economy Action Plan of the European Commission of 2015, upgraded in 2020, for developing sustainable jobs, growth, and investment and building a carbon-neutral, zero-waste and resource-efficient economy [20]. In fact, the long-shared linear economy model, based on the take–make–dispose paradigm has been used for decades to legitimize the indiscriminate harnessing of the limited natural resources, while mismanaging enormous volumes waste [21]. *Waste valorization* is a key-principle and a crucial driver towards a virtuous waste management system, zero-waste productions, and an ever-more circular concept, for the *green* as well as the *blue economy*.

Shells, as the principal mollusk-culture by-product, can find a legitimate space in this paradigm in several ways, independent from the waste producers, i.e., mollusk farmers and/or processing companies, restaurateurs, or consumers. For example, in Europe, mussels, clams and scallops are usually sold and served in full shells or processed and canned/frozen without shells, whereas oysters are commonly supplied to restaurants in

their full shells and consumed in half shells. By contrast, in Asia, mollusks are generally processed, and a major part of shells are removed at the point of harvest and regularly disposed of back into oceans or along coastlines [18].

To date, several options for valuable valorization opportunities have been reported. In the last four years, several reviews have been published regarding the valorization of mollusk shell waste, demonstrating the great and growing interest on this issue. Namely, in 2019, Jovic et al. [22] highlighted some potential applications of seashell waste to reduce the exploitation of natural limestone, comparing historical and modern shell reuses for applications such as soil conditioners, bioremediation agents, catalysts, and construction materials. Likewise, the review of Morris et al. [18] drew attention to shell waste as valuable biomaterials from both environmental and economical perspectives, underlining the fact that shell management represents the principal limitation of mollusk aquaculture development. In 2020, van der Schatte Olivier et al. [23] issued a review where the uses of shell waste were evaluated in terms of the ecosystem services provided by bivalve aquaculture. Their interesting results showed that global non-food services could cost up to USD 7.0 billion per year, including the reuse of shells. In 2021, Zhan et al. [24] and Alonso et al. [25] published two reviews on shell waste valorization, with the specific aim of including the reutilization pathways in the context of the circular economy and responsible aquaculture. More recently, Fraga-Corral et al. [26] focused their attention on the opportunity given by the reuse of aquaculture waste in Galicia, one of the main European aquaculture and fishery production sites, by including local aquaculture in the circular bio-economy paradigm. Medina Uzcátegui et al. [27] recently focused on sustainable alternatives to by-products derived from mussel processing.

On this same topic, the present review summarizes research from the last 22 years on to the reutilization of shell waste, i.e., applications in agriculture, construction, environmental protection, heterogeneous catalysis, and some new niche projects. All the examples reported aimed to map the principal opportunities of mollusk shells waste recovery, in order to implement a circular blue-economy model and provide more sustainable and responsible production systems. Moreover, a critical discussion of the effective sustainability assessment of shell waste reutilization was carried out.

2. Materials and Methods

The increasing opportunity to access to bibliometric data sources provides great opportunities for the development of widespread academic knowledge and research experiences in all scientific fields, including the field of shell waste valorization. In our study, publication data from 1990 to the present were retrieved from Google Scholar, including research articles, books and reviews, grouped by decade (Figure 1a) and published yearly since 2000 (Figure 1b). Approximately 14% of the results were published before 2000; this is the reason why our study focused on the following two decades and from 2020 to 2022. The keywords selected in the search of the available literature were 1) *mollusk shell, waste, valorization, mussel shell waste valorization, clam shell waste valorization, oyster shell waste valorization, scallop shell waste valorization*, without inserting values between any Boolean operator, and counting only once the publications listed at least twice.

It is worth noting the exponential increase in the number of scientific papers. The value of the three-year period 2020–2022 suffers due to the fact that only January and February are included for 2022. In Figure 1b, it can be observed that, in the first two months of 2022, the total amount of the publications for 2021 was surpassed. Perhaps it is no coincidence that only 2016 seems to deviate from the average trend as it was the year after the release of the Circular Economy Action Plan.

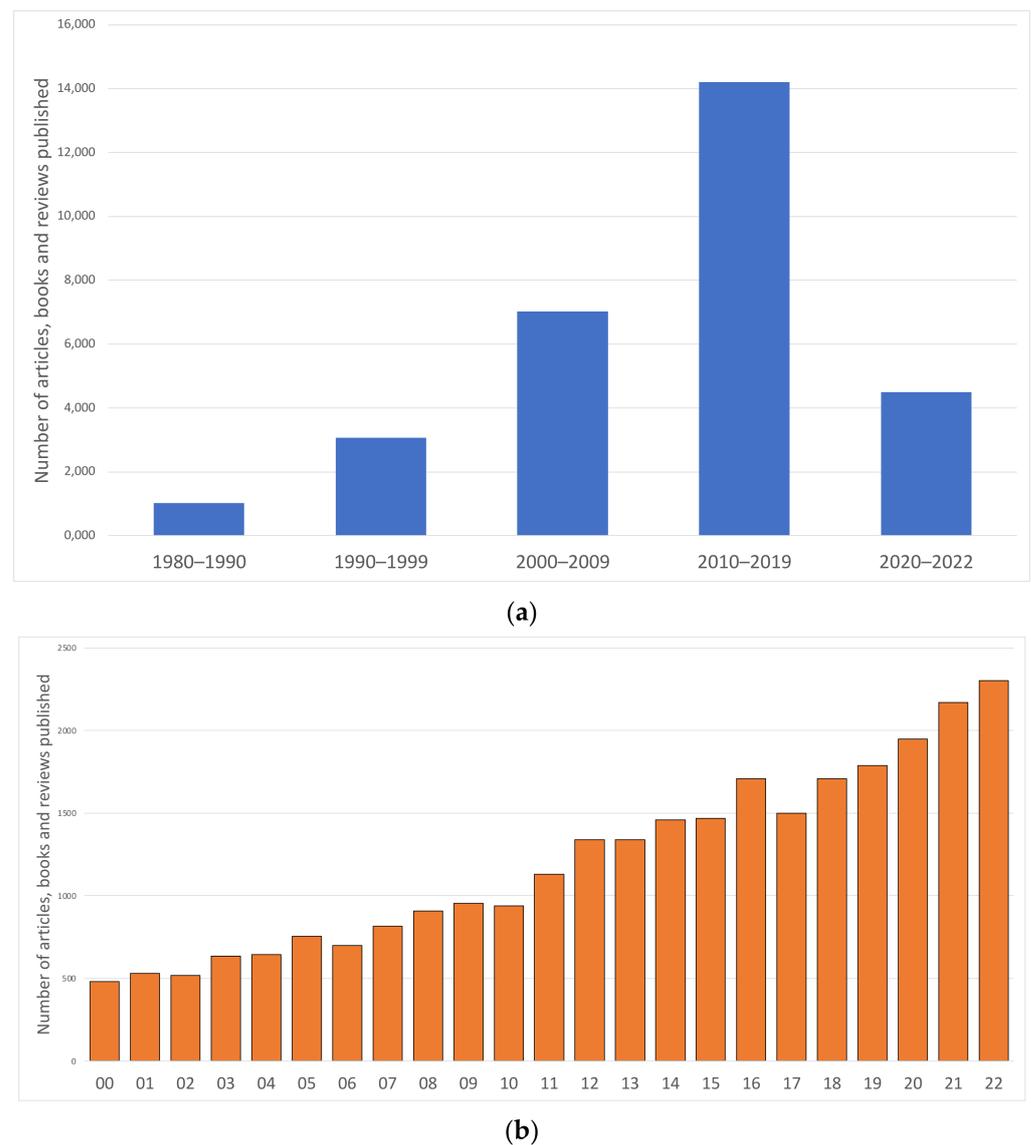


Figure 1. List of scientific publications in the field of mollusk shells valorization since 1980 grouped by decade (a) and by year since 2000 (b). In (b), the time interval on x-axis is expressed by omitting 20.

The following sections describe the results of the semi-systematic narrative review process of the literature, written in the English language, from internationally ranked journals or books [28], grouped in order to cover the different areas of applications, and selected by the criteria of number of reported citations, scientific quality and relevance of results.

Areas of valorization reported in literature since 2000 can be aggregated in some main groups, depending on the sector of utilization:

- In agriculture and livestock feed supplements;
- In soils, water, and wastewater remediation;
- In substitution of mined CaCO_3 in mortar and concrete;
- In heterogeneous catalysis for biodiesel production;
- As biofilters.

In addition, several small-scale and niche applications were recently proposed in the fields of cosmetics and biomedicines and for heavy metals removal from water and air [22].

Broadly speaking, the applications are enumerated in order of their industrial diffusion and applications.

The choice of how to conduct the literature search was the semi-systematic narrative review approach because it is a useful tool for providing an overview of shared issues and mapping the state of knowledge within a specific research field, as well as its theoretical perspectives; it can also investigate how research has progressively changed over time and developed over the years.

When using this approach to analyze the trend of published literature since 2000, it became clear that the older and well-established applications were focused on the agriculture, construction, and bioremediation sectors, representing about 90% of the overall literature. Notably, shell reuse in agriculture as soil conditioner was the most widely researched topic, covering more than 50% of the overall literature. In addition, it was found that about 70% of articles related to traditional applications of reuse were published before 2015, with an evident decline in more recent years due to the maturity reached by those sectors, which likely require a lower contribution of cutting-edge research. On the contrary, the use of shell waste as heterogeneous catalysts in biodiesel production has constantly increased; therefore, as expected, new applications in cosmetic and biomedicine have grown exponentially, increasing the quantity of references available by about 20 times over 20 years.

In the following sections, the principal fields of applications and valorization are briefly summarized.

3. Valorization in Agriculture and Livestock Feed Supplement

The older and principal market for shells waste lies in the agricultural sector involving the neutralization of acidic soils. Generally referred to as liming, this practice involves treating soil or water with lime (or a similar substance) in order to reduce acidity and improve fertility and oxygen levels. In acidic soils, shell waste seems to be a practical and interesting approach to suppress the needs in terms of calcium and provide good conditions for pH adjustments (i.e., liming), while promoting the goal of developing a circular economy and environmental sustainability [29,30]. Historically, shells from mussels (*Mytilus galloprovincialis*) were used after thermal treatment as a liming agent or as mulches for soil amendments in farming in Galicia [29]. Calcium carbonate (CaCO_3) is reported to neutralize acidic soil and increase soil organic matter, available P, and exchangeable cations concentrations, while improving its fertility and increasing oxygen levels [26].

Alvarez et al. [31] compared the effects of different types of mussel shell waste and commercial lime on soil and plant production. They found that the efficiency of treated mussel shell waste was comparable to commercial lime, with regard to the increase in pH, soil characteristics and improved pasture quality, increasing the Ca concentration in the plant tissues and favoring Ca and K absorption by plants. Lee et al. [32] carried out an environmental impact analysis based on a life cycle assessment of using oyster shell waste as a liming agent compared with the use of eggshell, demonstrating the major impact derived from electricity consumption on the calcination, milling and the drying processes.

Bivalves' shell waste has also been used as a liming agent, combined with other substances to improve soil fertility. For example, oyster shells were treated with coffee waste effluents and used as a soil fertilizer [33]. The high alkalinity, which contributes to a reduction in soil acidity, with the presence of coffee pulp, which provides soil fertility richness by means of high levels of organic matter, nitrogen, and potassium, makes it a valuable organic fertilizer. Furthermore, Kwon et al. [34] reported that blending 10–30% of powdered oyster shell blended with sewage sludge has positive effects on soil stabilization and fertilization. Paz-Ferreiro et al. [35] demonstrated an enhancement in soil chemical and biological parameters using mussel shells alone or in combination with cow slurry.

Moreover, crushed shells represent an important calcium supplementation (CaCO_3) when introduced into livestock feeding. Calcium supplementation is used to improve the health of livestock, particularly bone health, but also in laying birds as a supplement to improve the quality and strength of eggshells [36]. The replacement of the calcium present in limestone with that from oyster shells was proven to enhance bone development, egg production, and the strength, weight and thickness of eggs in hens [37–40] and ducks [41].

Another recent study found that oyster shells alone performed better than snail shells; wood ash or limestone, as a calcium supplement in terms of growth response; and weight gain and feed intake in laying hens [42].

According to the EU Regulation (EC) No 1069/2009, shell waste can be used as a feed supplement only if it is free from flesh, in order to be exempt from animal by-product classification [18]. The attribution of free-from-flesh standards is regulated by the respective authority of each member state. Eventually, for this type of valorization, the distance between shell production and farms must be taken into account from both an environmental and economic sustainability perspective [43].

4. Valorization in Contaminated Soils Remediation

The use of shell waste as amendments to remediate heavy metals in contaminated soil has attracted growing research interest and been widely applied [44,45]. Calcium carbonate increases soil pH, leading to the formation of calcium silicate hydrate and calcium aluminate hydrate, which create a relatively impermeable soil layer and decrease the mobility of heavy metals [46]. In addition, shells are promptly available and cost-effective materials, making them a desirable solution when vast amounts of contaminated soils must be remediated, including those in mining areas [47].

Oyster shell waste was applied for the treatment of cadmium- (Cd^{2+}) and lead (Pb^{2+})-contaminated soils near a closed mine. Moreover, an improvement of soil nutrients by applying oyster shell powder containing Ca^{2+} , Na^+ , Mg^{2+} and K^+ was reported [48].

Zhong et al. [49] reported the potential use of waste oyster shells as adsorbent and amendment agents for effective metal immobilization in both aquatic and sediment systems. Chen et al. [50] reported a significant reduction in arsenic (As^{3+}) leachability in highly contaminated soils when 2% oyster shell waste was applied in combination with 2% biochar, preventing shallow aquifer pollution, especially in sites with frequent precipitation. The use of 2% calcinated oyster shells was able to reduce the exchangeable Cd^{2+} in soil by 55%, and by 98% and 3% Cd^{2+} and As^{3+} content in the edible part of vegetable crops, making them safe for consumption [51]. A mixture of calcined oyster shells and waste cow bones was employed to remediate Pb^{2+} and copper (Cu^{2+}) in army firing range soils [52], reducing the availability of Cd^{2+} , Pb^{2+} , Cu^{2+} and zinc (Zn^{2+}) metal-contaminated soil [53].

In some cases, crushed and calcined mussel shells were used for soil remediation because they were rich in carbonates, oxides and different functional groups, with higher particle sizes and surface areas that could readily immobilize heavy metals [54]. Additionally, mussel shells could also increase soil pH and reduce the solubility of metals. Mussel shells drastically decreased Cu^{2+} , Cd^{2+} , Zn^{2+} and nickel (Ni^{2+}) mobility and the availability of plant uptake, reducing the overall risks of soil and water pollution [55–57].

The bioavailability of Pb^{2+} in military shooting range soils strongly decreased by 92.5% and 48.5% in mussel shells compared to the unamended soil, while decreasing the risk of ecotoxicity and supplying the soil with phosphorous [58].

5. Valorization as Biofilters in Water and Wastewater Treatment for Removing Metals

Adsorption has been reported as an attainable treatment process to eliminate traces of heavy metals from water, wastewater and landfill leachate [59,60]. Other conventional methods are currently widely employed, such as precipitation or ion exchange, but they are not always practical due to their high cost of operation and maintenance requirements, especially in developing countries [61]. Mollusk shells have recently been proposed as possible alternative biomaterials for heavy metal absorption [62]. They have been proposed for the treatment of electroplating wastewater from copper-mining industries in Guangxi of China [63]. Raw and acid-pretreated bivalves' shells demonstrated the efficient removal of Cu^{2+} through metal absorption based on the ion exchange between Cu^{2+} in the treated solution and Ca^{2+} from the shells. Nano-scale mollusk shell powders in aragonite were shown to effectively function as adsorbents in the treatment of Pb^{2+} -, Zn^{2+} - and Cd^{2+} -rich wastewater derived from plastics manufacturing, batteries, electroplating, metalworking,

and mining industries [64]. Oyster shells have been used for removing Cd^{2+} , chromium ($\text{Cr}^{3+/6+}$) and cobalt (Co^{2+}) from contaminated waters [65–67] and As^{3+} from tube well water [68] and mine tailings [69]. Unprocessed whole oyster shells have also been investigated for removing Cu^{2+} , Zn^{2+} , Cr^{6+} and Cd^{2+} in stormwater management infrastructure, with a good efficiency against Cu ions (80–95%), Cd ions (50–90%), Zn ions (30–80%), and hexavalent Cr (20–60%) [70]. Calcinated and raw mussel shells showed a high capacity of mercury (Hg^{2+}) retention [71] and were recently studied for phosphate removal from water, yielding very promising results: above 90% [72–74].

Lately, mollusks shells have also been used in combination with waste glass as absorbents for removing the direct blue 15 azo dye [75,76], methylene blue [77], and red dye [78] from industrial wastewater. Moreover, they have been proposed as absorbents for the removal of emerging contaminants, such as hormones, personal care products, pharmaceutical products and pesticides, from domestic and industrial wastewaters, obtaining efficiency values higher than 30% [79]. There were some studies from recent publications where shells waste were recommended for their removal of rifampicin antibiotic [80] and biocides residues from waters, such as triarylmethane [81]. Powdered mussel shells were used for the removal of methyl blue and methyl red, together with Cr^{6+} , Cd^{2+} and Cu^{2+} from wastewater, demonstrating a capacity of almost 100% removal, as well as improving the settling properties of the activated sludge flocs [82].

6. Valorization as a Substitute for Mined CaCO_3 in Mortar and Concrete

Concrete—a mixture of gravel, sand, cement, water, and mortar formed by cement and sand—is among the most widely used building material. The concrete and mortar industry has harmful effects on the environment, especially in terms of greenhouse gas emissions. The use of mollusk shell waste as a substitute for concrete and mortar ingredients is attracting research interest as a viable alternative that may reduce our dependency on conventional components [83,84]. In order to evaluate the possible application of bivalves' shell in the production of calcitic lime, Ferraz et al. [85] characterized shells of eight different species of bivalves for their mineralogical, chemical and thermal properties, compared with commercial limestone as reference. It was noted that scallop and oyster shells are composed mainly by calcite, similar to the reference limestone, whereas mussel shells mainly contain calcite. Additionally, aragonite, edible cockle, wedge, razor clam, dog cockle and clam shells contained aragonite as principal component, with minor or negligible amounts of calcite.

Researchers generally suggested that a few important steps of treatment should be considered prior to reusing seashell waste: washing, calcining, and crushing to the desired size. As seashells are primarily obtained as waste, the proper handling and treatment of the waste must be carried out before they are incorporated in concrete, to ensure the removal of impurities. Additionally, heating at 500–650 °C is required to remove the organic matter attached to the seashell, and crushing is needed to ensure better bonding between the seashell aggregate and the cement. Several studies already investigated the opportunity of integrating bivalve shells in ordinary [86–89] and pervious [90–92] concrete, mortar [93–96], air lime mortar [97,98], artificial stone [99,100], and bricks [101–103], in a broad range from 5 to 90% [104–107]. However, generally speaking, it was reported that the inclusion of bivalve waste as an aggregate in mortar or concrete reduced its strength and workability properties by more than 20% [108]. Due to these difficulties and the strict safety regulations in building materials, the wide-scale application of mollusk shells in construction is not yet an established market [18]. Alternatively, there are many historical examples of shells in residential houses or walls in different coastal areas [109].

7. Valorization as a Catalyst in Biodiesel Production

Being rich in calcium carbonate, when calcined at a proper temperature, shells are converted into the metal oxide CaO, which can be used as a heterogeneous catalyst in the transesterification process of biodiesel production [110]. Chemically, biodiesel contains

alkyl esters of long-chain fatty acids derived from renewable feedstocks, such as vegetable oils and animal fats [111]. Transesterification with methanol is one of the most utilized routes for first-generation biodiesel production, consisting of a chemical reaction of vegetable oil/animal fat with alcohol in the presence of a base catalyst to form glycerol and esters [112]. Even though homogeneously catalyzed biodiesel production processes are relatively faster and show higher conversions, heterogeneous catalysis using CaO is becoming more popular because it overcomes some disadvantages of the traditional method, namely side-reactions leading to soap production, and the difficulty of separating the catalyst from products at the end of the reaction, thus leading to high costs [113]. Hu et al. [114] showed how calcinated and rehydrated mussel shells could be used as catalysts for palm oil transesterification in five tests of reuse with a yield of 95% in the first test and 59.1% in the last test. The negative effects of the reusability of shells on biodiesel yield were also confirmed by Rezaei et al. [115]. Mussel shells were used as catalysts in transesterification, achieving the following yields for a number of substances: methanol of palm oil, nearly 95% yield [116,117]; castor oil, 91.17% yield [118]; sunflower oil, 90–97% yield [119]; jatropha oil, 97.54% yield [120]; and peanuts and rapeseed oil, 94 and 96% yield, respectively [121].

Biodiesel from camelina oil was obtained using mussel, clam and oyster shells with 95, 93 and 91% yields, respectively [122].

Clam shells were used in both first-generation biodiesel from rapeseed [123,124], jatropha [125] or cottonseed [126] oils and second-generation biodiesel production in the transesterification of waste frying oil, with a conversion of 94.25% compared with 67.57% obtained using commercial CaO [127].

KI-impregnated and calcinated oyster shells were used as solid catalysts for the transesterification of soybean oil with an 86% yield [128]. Combusted oyster shells were exploited in soybean oil transesterification under the optimum reaction conditions and reached a 73.8% yield with a high biodiesel purity (98.4 wt.%) [129]. Crude bio-oil was extracted from the seeds of capers and transesterified into biodiesel using CaO from oyster shells [130]. Oyster shells were also employed for biodiesel synthesis from waste cooking oils, achieving a 87.3% yield [131].

8. Niche Applications

This section is dedicated to the potential applications of shells waste, which are currently being tested only at laboratory level or have not been optimized on a large scale. Consequently, economic or environmental sustainability analyses for these applications have not yet been conducted. However, there are some possible innovative uses for shell waste that are worth highlighting, i.e., biomedical, and cosmetic applications, as well as uses as additives for biocomposite polyesters, de-icer grit, or green roofing substrates.

In recent years, the potential biomedical utilization of mollusk shells as natural sources of calcium has attracted growing interest due to the similarity of the bio-calcification process in mollusks and osteogenesis in the human body [132]. For example, oyster shells are known as one of the few active surrogates for calcium insufficiency in osteoporosis treatment [133–135]. Bone is an inorganic matrix formed by 60–70% of hydroxyapatite, a calcium phosphate mineral, which is bioactive and biodegradable [136]. Bone is one of the main implanted tissues in the human body, and bone defects caused by fractures, diseases or genetic disorders are considered as one of the main burdens for healthcare systems worldwide [137]. The research on innovative and biocompatible scaffolds for regenerating damaged bones is paramount. Nowadays, polycaprolactone polymer is considered the most promising materials due to its low toxicity, low immune reactivity and good mechanical properties [138]. Recently, oyster shell powder was proposed in combination with polycaprolactone as a composite scaffold to overcome its low bioactivity [139]. Hydroxyapatite derived from clam [140,141] and mussel [142,143] shells was also demonstrated to be useful for bone-related applications.

Hydroxyapatite from powder clam shells was used as an additive ingredient in the formulation of emulsions, such as sunscreen lotion, by the substitution of oxybenzene, zinc

oxide and titanium dioxide [144,145]. Clams, mussels and oysters shells have also been studied as potential sources of collagen for cosmetic applications [146].

Powdered and calcinated bivalve shells were used as additives for polyesters, namely polylactic acid (PLA), in order to endow antibacterial properties to biocomposites [147]. Antibacterial properties resulted from the presence of hydroxyl ions and active oxygen species generated at the shells surface, which inactivated the microorganisms [148]. Mussels shells were valorized as a filler for compostable co-polymers made of PLA and polybutylene adipate terephthalate (PBAT) [149].

Bivalve shells were successfully applied in the US in road de-icing processes as an alternative to sodium chloride, which is known to lead to severe water pollution due to salt runoff occurring in late winter and spring [150]. Another potential use of mollusk shells is in green roofing structures, such as drainage layers, or to help with acidic rain neutralization [18,151]

Oyster shells and giant clam shells have been also proposed as promising ecological and economic approaches to producing artificial coral reefs. Reef rocks are regularly and indiscriminately harvested from the wild to supply the marine aquarium industry, becoming one of the most pressured and degraded marine ecosystem [152]. Replacing the natural coral skeleton and coral algae with oyster shells was demonstrated to help provide the essential substrates for bacterial communities that mediate nitrogen cycles and host an enormous amount of other natural organisms, such as anemones, polychaetes, echinoderms and many other invertebrates [153]. Examples of oyster reefs can be seen across the US Atlantic, including Chesapeake Bay, and the southeastern gulf coasts of the US [154].

9. Discussion of Effective Sustainability Perspectives

The above-mentioned applications summarize some of the key scientific literature on shells as a natural calcium carbonate source. Bivalve mollusk culture represents an essential part of global aquaculture, which is a crucial aspect of the future of building food security. The large amount of waste produced by this industry in the form of shells is currently underutilized or dumped in landfill, causing several environmental problems, i.e., bad smells, damage to natural landscapes, and sanitation problems, even though we report that they might become a valuable resource in various fields.

Even though our recurrent key focus is the circular economy and sustainability assessment of shell valorization, it is worth noting that the literature is often limited to reporting a list of potential applications and hardly ever deals with the issue of the effective sustainability of various forms of reuse in a quantitative way. For example, only in Morris et al. [18] was the issue of the distance between shell source and processing facility addressed, citing the research article of de Alvarenga et al. [43]. A distance of 323 km was determined as a limit beyond which the environmental benefits of oyster shells reuse, would be lost over landfill disposal, calculated for a Brazilian case study by means of a life cycle assessment (LCA).

Although shell waste is an abundant and inexpensive natural renewable resource, before their application on large-scale, some obstacles should be overcome, or at least put in evidence, for an overall sustainability analysis. Shells could be recycled because of their high content of CaCO_3 , but when discarded after production or food consumption, the organic residues and other impurities must be removed by calcination at high temperatures followed by powdering and sorting processes to reach a CaCO_3 purity of 90–95%, depending on their final use [155]. Moreover, in the context of a circular economy, the potential impact of shell waste collection and management systems should not be neglected in an overall sustainability analysis.

In fact, based on the current habits of worldwide consumers, most bivalves are predominantly consumed as fresh and at home, which makes the collection of the shells difficult [25]. This represents the principal obstacle present in any of the applications proposed above, but it is rarely taken into account and discussed [18].

In other words, what in theory appears to be a sustainable solution because it traces a virtuous route of valorization, often clashes with a different reality, where the environmental impact of waste transport and waste treatment can have dramatic effects.

An overall sustainability assessment from a circular economy perspective cannot neglect these fundamental aspects.

As an example, in Italy, about 100,000 tons of mollusks are sold per year, most of them for domestic consumption (about 80%). About 90% of these mollusks are provided by national production; the rest are imported by other European countries, such as Spain and France [156].

Overall, considering that shells represent on average 80% of fresh weight, about 80,000 tons of shells waste is produced annually. In practice, with a national average percentage of waste selection of about 60% [157], 50,000 tons of shells waste could theoretically enter into valorization routes each year. It can be assumed that the municipal system of door-to-door separate waste collection used for organic fractions of waste could also be shared with shells collections. Therefore, any net additional environmental impact would be a burden on the sole shells collection, and the distance between thousands of collection points (houses and restaurants) and the waste hub would not be counted in the overall sustainability assessment. Based on a life cycle assessment (LCA) of shell valorization carried out by Iribarren et al. [155], the CO₂ eq. emissions for processing 100 tons of shells for CaCO₃ were evaluated as 0.775 tons, which is surprisingly higher than an end-of-life scenario in landfill (quantified in the aforementioned article as 0.496 tons CO₂ eq./tons of mussel shells). Although the landfilling option accounted for a more apparently favorable impact, this alternative is totally discouraged, representing the worst choice in terms of sustainability and the valuable exploitation of waste. On the other hand, de Alvarenga et al. [43] and Lee et al. [32] demonstrated a considerable environmental benefit related to CaCO₃ recovery, producing an impact of about 40% or less compared with landfilling or incineration as undifferentiated waste.

Assuming that 57 g CO₂ eq/ton km is emitted by a large truck during EU road transport [158], to match the total CO₂ eq. emissions from the processing of the theoretical 50,000 tons of shell waste in Italy (38,750 tons CO₂ eq/year, based on Iribarren et al. [155]), the average distance between the processing facilities and the target-site of shell-derived CaCO₃ can be estimated as approximately 14 km.

Apart from studies by researchers, the cooperation and financial support from local governments and related industries are obviously indispensable for develop industry-scale and high-value-added reutilizations, thereby promoting sustainable shellfish aquaculture. Clearly, the overall sustainability of any shell reutilization must be evaluated in terms of both environmental and economic impacts, and the resulting economic value must be considered. Therefore, the distance between shell production and shell utilization sites should be carefully evaluated, as well as the potential need of storage costs, in view of a real application in a circular economy. The recycling and potential applications of shells have been the focus of several scientific articles over the last twenty years, some of which are well-established and already widely references; others remain at the laboratory scale as niche applications that are not yet fully explored. An increasing global interest points to a great opportunity for the utilization of shell waste and can contribute to overcoming one of the principal hinderances to bivalve aquaculture development as sustainable food production worldwide. There is now a real opportunity for interdisciplinary researchers and the industry to work together to enable bivalves to become a greater part of the circular economy paradigm, through the reuse and valorization of shells.

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