Cleaner Engineering and Technology 20 (2024) 100743

Contents lists available at ScienceDirect

Cleaner Engineering and Technology

journal homepage: www.sciencedirect.com/journal/cleaner-engineering-and-technology

Preliminary study on the application of waste bivalve shells as biofiller for the production of asphalt concrete

L. Caroscio [a], B. De Pascale [a,*], P. Tataranni [a], C. Chiavetta [b], C. Lantieri [a], A. Bonoli [a]

[a] Department of Civil, Chemical, Environmental and Materials Engineering-University of Bologna, Italy
[b] ENEA - Italian National Agency for New Technologies, Energy and Sustainable Economic Development, Italy

ARTICLE INFO

Keywords:
Asphalt concrete
Biowaste filler
Bivalve shells waste
Oyster shells
Mussel shells
Clam shells

ABSTRACT

The shells of molluscs are a common by-product of the aquaculture industry, and their management represents a significant environmental challenge. Although mollusc farming is considered a low-impact food production, improper shell management could make bivalve farming less environmentally efficient. To address this issue, research is exploring new approaches to reduce waste accumulation and convert shell waste into a valuable resource. The shells of bivalves are functional materials from biological waste, composed mainly of $CaCO_3$, and can be used as secondary raw materials in various applications. In order to meet the demanding environmental target, the road sector is increasing the use of recycled materials in new construction or maintenance of old ones. The present work illustrates the results of several laboratory tests carried out to determine the physical and chemical properties of three different crushed bivalve shells waste for the application as filler in asphalt concretes. The present study highlighted the similarity of these materials with the limestone filler since no significant discrepancy between the mechanical (or technical) performance of the biofiller and the traditional limestone filler are detected through the test carried out, promoting their use in new asphalt concrete mixtures.

1. Introduction

Mussel shells have a mineralogical composition characterized by about 95 percent by mass of calcium carbonate usually in the crystallographic form of calcite or aragonite (Currey, 1999). Because of this peculiarity, they have been studied for several years for use as a secondary raw material (Morris et al., 2019). This approach allows avoiding large quantities of shells, approximately 13 million tons produced annually by bivalve mollusk aquaculture, from ending up in landfills, instead favoring a virtuous cycle that complies with European directives on waste management (Regulation, 2009; Jović et al., 2019). These directives promote the adoption of recycling and reuse practices for raw materials, rather than their disposal in landfills or incineration.

In 2021, in the EU Member States, 552,669 tons of bivalves and other aquatic mollusks and invertebrates were farmed (The EU fish market,). Since oysters, mussels, and clams accounted for over 99% of the volume and value of the total aquaculture production of this group of products in the EU, it was considered to focus this study on the valorization of these three types of bivalve shells.

One of the main problems of bivalve farming is related to the ratio between product and edible portion and the amount of waste produced; for every kilogram of fresh bivalve product, a percentage ranging from 65 to 90% is represented by their shells (Cala et al., 2023; The State of World Fisheries, 2022).

The disposal of shells poses significant challenges for those involved in the shellfish industry, impacting both practical and economic aspects (Seesanong et al., 2022; Azra et al., 2021). Huge amounts of bivalve shell waste are often landfilled or dumped into the sea, causing ecological impacts due to organic debris attached to the shells and anoxia of the seabed (Summa et al., 2022). To cope with this crucial situation, alternative approaches for recycling bivalve shells are currently investigated (Diokhane et al., 2022; Sakr et al., 2022). In fact, mussel shells, managed as by-products of animal origin, have found applications as a dietary supplement for poultry farming and as agricultural applications (Çatli et al., 2012; Oso et al., 2011; Osorio-López et al., 2014; Directive 2008, 2008). In recent years, research has been directed towards experimental uses such as those related to design or the creation of artificial reefs for marine habitat restoration (Mathews et al., 2023; Yoris-Nobile et al., 2023; Corbau et al., 2023).

Regarding the use of shells as a secondary raw material within construction materials, shells have been studied for about 20 years (Eziefula et al., 2018). Since many building materials are subject to strict

---

[*] Corresponding author.
E-mail address: beatrice.depascale3@unibo.it (B. De Pascale).

https://doi.org/10.1016/j.clet.2024.100743
Received 5 February 2024; Received in revised form 22 March 2024; Accepted 31 March 2024
Available online 4 April 2024

regulations to ensure performance and safety, as indicated in EU Regulation No. 305/2011, it is necessary to pay attention to permitted uses (Regulation, 2011). Much research has been conducted on the integration of shells into cementitious materials to minimize excessive dependence on natural resources (Tayeh et al., 2019; Martínez-García et al., 2017). The shells used do not exclusively come from bivalve mollusks but also from gastropod mollusks, although in the latter case, the quantity of waste available is lower (Shetty et al., 2023).

Still, a limited number of studies have investigated the use of bivalve shells in asphalt concrete as coarse aggregates or filler (Çevrim and Iskender, 2022; Alharthai et al., 2021). Some of them have explored the impact of utilizing waste seashells as modified materials in asphalt concrete pavement (Fan et al., 2022; Wu et al., 2022). In the literature few studies investigated the use of seashells as biofiller in asphalt concrete mixture analysing the effect on asphalt-aggregate interfaces based on thermodynamic properties (Lv et al., 2021; Guo et al., 2021).

Fillers in asphalt concrete (AC) represent the finest particles of the lithic skeleton of the mixture. Thanks to its powdery composition, filler confers specific properties to bituminous and non-bituminous mixtures (Targino Bezerra et al., 2011). Filler exhibits a grading curve wherein over 75% of the particles passes through the 0.063 mm sieve (BSI Standards Publication Aggregates for, 2013). It can deeply affect the properties of asphalt concrete such as strength, air voids content, thermal susceptibility, and plasticity (Antunes et al., 2017; Lagos-Varas et al., 2020). In general, the mineral filler in asphalt mixture should fulfil certain requisites, including adequate coating ability, inability to chemically react with binding agents and certainty of good adhesiveness with the binding agent (Arabani et al., 2015). Furthermore, mineral filler is mixed with asphalt and forms a mastic that contributes to the rutting and cracking properties of ACs (Tiwari et al., 2023; Choudhary et al., 2021). These properties are generally common for traditional fillers such as calcium carbonate powder, limestone filler, and Portland cement (Ayunaning et al., 2022). With the development of the concept of environmental sustainability and the increasing awareness about the responsible use of natural resources and recycling waste materials, over the past 25 years, numerous researches have been undertaken to substitute natural components of asphalt mixtures with waste and bio-waste products (Sangiorgi et al., 2016; Modupe et al., 2021; De Pascale et al., 2023). This study intends to investigate and evaluate the possible replacement of mineral filler in asphalt concrete mixtures with crushed waste bivalve shells. Precisely crushed mussels, oyster and clam shells bio fillers are included in the study. In order to be used as filler, the bivalve shells underwent a pre-treatment aimed at removing impurities and organic materials, and a mechanical treatment for obtaining the proper size of the particles typical of a filler material. All the tests undertaken followed the European standard EN 13043, which establishes the properties of aggregates and filler being used in asphalt concrete mixtures.

2. Materials and methods

2.1. Materials

The aim of the present research is to evaluate the feasibility of three different waste coming from the shells of bivalves to be used in the production of bituminous mixtures as bio fillers. The waste shells are collected from aquaculture farms situated in Italy, specifically in La Spezia for mussels and oyster, and in Goro for clams. The bivalve wastes are transformed, in laboratory, into powdery materials and are reported in Fig. 1.

The three bio fillers derived from the crushing and milling of waste bivalve shells come from oyster shells, clam shells and mussel shells. Before being transformed into powdery material, the waste shells need to undergo some pre-treatment in order to remove the impurities and organic residues. The pre-treatment process encompassed two main steps: the first step involved a manual cleaning of the shells and a subsequent period of 1 h in oven a 400 °C to remove the remaining organic compounds. Then the second step concerns the crushing and milling of the bivalve shells into the final materials with the desired size distribution using two different mills.

Bivalve shells are mainly composed of calcium carbonate ($CaCO_3$), which correspond to the composition of the traditional limestone filler used in Italy to produce asphalt concretes. The limestone filler comes from Ancona, Italy and is commonly employed in the production of hot mix asphalt. Different tests were carried out in order to verify the properties of the biofillers and their possible compatibility with bitumen. To develop the test on bituminous mastics, a traditional 50/70 pen bitumen is used and the rheological properties are reported below in Table 1.

2.2. Test methods

Based on the reference standard for bituminous materials (EN 13034), an experimental program was organized to investigate the chemical and physical properties of the filler, considering the following laboratory tests:

- Size distribution (EN 13043);
- Determination of the organic material (ASTM D2974);
- Water content (EN 1097-5);

Table 1
Rheological properties of neat bitumen.

Test	Unit	Value	Standard
Penetration 25 °C	dmm	53	EN 1426
Softening point	°C	50	EN 1427
Dynamic viscosity 60 °C	Pa.s	327	EN 13702

Fig. 1. Oyster biofiller, clam biofiller, mussel biofiller.

- Particle density (EN 1097-7);
- Voids of dry compacted filler (EN 1097-4);
- Variation in “Ring and Ball” temperature (EN 13179-1);
- Harmful fine (EN 933-9);
- Water solubility (EN 1744-1);
- Determination of carbonate content (EN 196-2).

The experimental program encompassed some of the traditional tests used to assess the properties of filler used in bituminous mixtures. These tests are considered, by the literature, as essential to investigate and describe the qualitative properties of fillers (Sangiorgi et al., 2017; Russo et al., 2022).

3. Results and discussions

3.1. Geometrical and physical characteristics

3.1.1. Size distribution

In Table 2 the passing percentage at sieve 63 μm is reported. With these percentages, the size distribution of the filler is presented, and it is in compliance with the standard EN 13043, following a standard sieving procedure. The three biofillers are named Biofiller M for the material produced by mussel shells, Biofiller O for the one produced by the oyster shells and Biofiller C for the filler produced by the clam shells.

Based on the European standard, the filler used in asphalt mixtures must have 70% of particles pass through a 63 μm sieve. Each of the tested biofillers meets the requirements of the standards. As confirmed by various studies, particles size and their shapes significantly influence the mechanical properties of the mastic, particularly in this case, some difference in the particle dimension is detected if compared with the traditional limestone filler but still in compliance with the technical standard (Topini et al., 2018).

3.1.2. Determination of the organic material

This test covers the measurement of organic material present in the biofillers. The bivalves’ shells were pre-treated in order to remove the majority of the impurities before being transformed and crushed into the final bio fillers. The organic content is then tested to assess how much of organic compound is still present and how effective the pre-treatment process was. The organic material is determined by igniting the oven-dried specimens in a furnace at 440 °C, in compliance with the ASTM D2974 standard. The results presented here (Table 3) are average values obtained from three repetitions for each filler.

From the current results it can observed that the pre-treatment process almost completely eliminated all the organic compound present on the shell and the three bio fillers have an organic content nearly equal to zero and in line with the results obtained for the limestone one.

3.1.3. Water content

Moisture may originate from either the particle surfaces or the available pores within the powdery material. The total mass of free water within the three biofillers is tested trough the ventilated oven method drying test in accordance with the European standard EN 1097-5.

The water content of the filler is determined by calculating the variance between the humid mass (or initial mass) and its dried mass, expressed as a percentage of the dried mass. Actually, this dried mass represents the constant weight of the test specimen after undergoing oven drying. Table 4 shows the results obtained for each biofiller.

Table 3
Percentage of organic materials.

ASTM D2974	O_m [%]
Limestone Filler	0.00
Biofiller M	0.45
Biofiller O	0.70
Biofiller C	0.24

The water content of Biofiller M, Biofiller O and Biofiller C are totally comparable to the value obtained for the limestone filler.

3.1.4. Particle density

The particles densities of the biofillers were calculated using a pycnometer. The test sample was prepared with a minimum mass of 50 g, following EN 932-2 standard, and subjected to drying at 110 °C until a stable weight was reached. The dried fillers were sieved through the 0.125 mm mesh and the sieved fraction evaluated. In Table 5 are reported the particle densities of the three biofillers.

The density value of the three recycled fillers are nearly equivalent to the density of the limestone filler, as shown by the results. The slightly difference between each material are mainly due to the small difference in terms of chemical composition of each filler. The bivalve shells are mainly composed of calcium carbonate and a smaller percentage is represented by calcium phosphate and conchiolin, a specific protein produced by the mollusc.

3.1.5. Voids of dry compacted filler

Rigden voids, also known as intergranular porosity refer to the air-filled space within the material, expressed as a percentage of total volume of the filler. The test can be applied to natural, artificial or recycled filler and it consists of compacting the filler through a normalized method.

The percentage of intergranular voids is determined from the initial weight of the specimen, the density and the weight of the specimen compacted after a specific number of strokes. Through the Rigden device, the intergranular voids are determined mechanically. This property of the fillers plays a crucial role in shaping the physical and mechanical performances of the bituminous mixture, impacting the stability of the bitumen (Jyothi et al., 2018). Table 6 reports the Rigden voids percentage of each biofiller.

The current results provide insights into the potential quantity of bitumen that biofillers can adsorb and the stiffening capacity these materials impart on the bituminous mastic. This test facilitates the determination of the remaining amount of bitumen, specifically referring to the bitumen that does not get adsorbed by the voids in the particles. This serves as a crucial indicator that helps in the assessment of the rheological behaviour of the mixture. It has emerged from different studies that a low value of filler porosity implies a lower rigidity of the mastic due to the fact that the filler particles are not anymore in close contact (Ruiz et al., 2020; Sangiorgi et al., 2014).

While the results reported in Table 6 may differ somewhat from those of the limestone filler, the values are in accordance with the European standard EN 13043, that restricts the suggested Rigden Voids values for fillers suitable for bituminous materials in the range 28-55%.

Table 2
Material passing at sieve 63 μm.

EN 13043	P63 [%]
Limestone Filler	94.2
Biofiller M	80.3
Biofiller O	82.2
Biofiller C	79.4

Table 4
Water content (EN 1097-5).

EN 1097-5	Initial Mass [g]	Dried Mass [g]	Water content [%]
Limestone Filler	53.83	53.72	0.20
Biofiller M	201.9	201.81	0.33
Biofiller O	199.15	199.08	0.31
Biofiller C	202.9	202.85	0.27

Table 5
Particle density (EN 1097-7).

EN 1097-7	Particle Density [Mg/m^3]
Limestone Filler	2.70
Biofiller M	2.68
Biofiller O	2.63
Biofiller C	2.64

Table 6
Rigden voids (EN 1097-4).

EN 1097-4	Weight of compacted filler [g]	Density of filler [Mg/m^3]	Height of compacted filler [mm]	Rigden Voids [%]
Limestone Filler	9.95	2.70	12.37	41.00
Biofiller M	9.68	2.68	10.83	33.26
Biofiller O	9.51	2.63	11.17	35.14
Biofiller C	9.59	2.64	11.17	34.84

3.1.6. *Variation in "Ring and Ball" temperature*

The EN 13179-1 standard proposes various tests to evaluate the interaction between the bitumen and the filler. The change of the temperature of the bitumen is determined using the "Ring and Ball" method after incorporating the fillers and creating the mastic. The proportions, in volume, for creating the mastic are:

- 61.2% of limestone filler and 38.8 of bitumen;
- 60.6% of Biofiller O and 39.4% of bitumen;
- 61.1% of Biofiller M and 38.9% of bitumen;
- 60.7% of Biofiller C and 39.3% of bitumen.

Only filler with particle of less than 125 μm can be used for creating the mastic. The test involves the evaluation of the average temperature at which the two samples, with and without the addition of filler, soften. A steel sphere is place on top of the samples, and at the softening point it touches a metal plate underneath at a standardized distance of 25 (±0.4) mm. Through the calculation of the difference between the two average temperature the Ring and Ball delta is known.

The rheological characteristics of bituminous mastic can be significantly influenced by the physical properties of the biofillers used. Additionally, the compatibility between the filler and bitumen at a chemical level is another aspect that affects the overall properties of the mastic (Sholichin and Ayunaning, 2019).

The results shown in Table 7 report value from 2 to 5 °C higher than the variation of temperature detected for limestone filler.

As stipulated in EN 13043 for conventional fillers, the Ring and Ball delta should fall between 8 and 16 °C. Additionally, certain studies emphasized that the bituminous mixture performances are improved when a delta Ring and Ball is between 12 and 16 °C (Bamigboye et al., 2021; Nciri et al., 2018).

Table 7
Variation in Ring and Ball temperature (EN 13179-1).

EN 13179-1	Softening T° bitumen [°C]	Softening T° bitumen + filler [°C]	Variation Ring and Ball T° [°C]
Limestone Filler	47.5	59.5	8.0
Biofiller M	49.9	62.8	13.0
Biofiller O	49.4	63.3	13.9
Biofiller C	49.4	59.7	10.3

3.2. *Chemical composition*

3.2.1. *Harmful fine particles (Methylene Blue Test)*

In the EN 933-9 Standard is described the Methylene Blue Test method that consist in adding a drop of methylene blue of 10 g/L of concentration at regular intervals to a solution of filler and water. The water with filler suspension is inspected each time a drop of methylene blue is added, and the stain and halo produced on a filter paper is evaluated. The aim of the test is to assess the presence of fine particles within the filler, usually composed by hydrated aluminosilicate clay mineral. The fine fraction is absorbed by the methylene blue in an aqueous solution, facilitated by its surface charge and cationic capacity. The absorption of methylene blue solution increases proportionally with the amount of fine particles present in the biofillers. The presence of these particles can reduce the filler-bitumen compatibility The methylene blue values are reported in Table 8.

These results are similar for the three bio fillers and in line with the value founded for the limestone one.

3.2.2. *Water solubility*

In accordance with EN 1744-1 Standard, the water solubility of the filler test sample is determined by extracting the dried filler with a predetermined volume of water equivalent to fifty times the mass of the filler itself. The three biofillers are free of any added calcium hydroxide, so based on the standard, the water solubility can be tested with the standardized procedure. Two repetitions are performed for each filler. A glass bottle is used and filled with the fixed quantity of dried material and distilled water. The bottle with the solution is carefully sealed avoiding the contamination with other materials and placed on a mechanical shaking device which mixes and rollers the specimen for 24 h to avoid the sedimentation of the bio filler. After mixing, with a funnel and a filter paper all the liquid is filtered and then placed in an oven until the solid residue is dried and reaches a constant weight. The results of the Water solubility test are reported below in Table 9.

EN 13043 suggests that the water solubility value for filler to be used in asphalt mixture should remain below 10%. Even if the value obtained for the three biofiller are much higher respect to the limestone one, the results still comply with the limit suggested by the European Standard.

3.2.3. *Determination of carbonate content*

Following EN 196-2 Standard, the determination of carbonate content was evaluated with a Dietrich Fruhling calcimeter. Two samples for each biofillers were tested. The specimens are placed in a glass container, in which a test tube containing about 2/3 of hydrochloric acid (HCl) and 1/3 of water is carefully added in it. The HCl and H_2O solution is then poured inside the glass container and shaken to start the reaction with the filler. The CO_2 developed by the reaction moves the water column of the device and then the stabilized height of the water can be assessed. Based on the quantity of CO_2 that is produced it is possible to detect the amount of $CaCO_3$ inside the tested bio filler. Two samples, each weighing 10 g, were examined for each material and Table 10 reports the results of the carbonate content of each bio fillers.

From the determination of carbonate content is it possible to state that Biofiller O contains a high percentage of carbonate content, almost equal to that of pure calcium carbonate. The calcimetry test, however

Table 8
Methylene blue test value (EN 933-9).

EN 933-9	Mass of the sample [g]	Methylene blue added [mL]	Methylene blue value [g MB/kg Filler]
Limestone Filler	30	10	3.3
Biofiller M	30	12	4
Biofiller O	30	9	3
Biofiller C	30	9	3

Table 9
Water solubility (EN 1744-1).

EN 1744-1	Mass filler before extraction [g]	Mass filler after extraction [g]	Water Solubility [%]
Limestone Filler	5.02	5.00	0.50
Biofiller M	5.02	4.58	8.77
Biofiller O	5.02	4.80	4.48
Biofiller C	5.01	4.66	7.08

Table 10
Calcium carbonate content.

EN 196-2	$CaCO_3$ [%]
Limestone Filler	98.0
Biofiller M	90.3
Biofiller O	98.3
Biofiller C	94.8

cannot identify the mineral form in which the calcium carbonate is present and which are the others constituents of the biofillers. It is worth noting that this test is not mentioned in the EN 13034 Standard. However, considering the well-known chemical compatibility of calcium carbonate with bitumen, this method can be regarded as an indirect indicator of the possible affinity of the tested biofillers with bituminous binders.

In summary, the size distribution of fillers in asphalt concrete can be tailored through specific mechanical processes, offering flexibility in achieving desired grading sizes. Previsous studies examining various filler materials consistently report low water content values, typically below 1.5%, highlighting the importance of moisture control in asphalt mixtures (Moustafa et al., 2017; Choudhary et al., 2022). The compliance of all biofillers with standard ranges, particularly Biofiller O and Biofiller M aligning with prior research, underscores their potential suitability for asphalt applications. The high water solubility associated with these biomaterials is attributed to their natural composition. Exploring novel materials and methodologies in asphalt concrete research is crucial for optimizing performance and sustainability.

4. Conclusions

The results of the tests carried out in the experimental program following the EN 13043 are discussed and commented in the present paragraph.

The European standard sets the condition and characteristics of traditional fillers to be used in asphalt concrete but does not underline any constrains and restriction for recycled and artificial fillers. Nonetheless, the limit imposed by the European standard does not exclude the possibility of using the tested fillers, but they just give ranges for obtaining a proper asphalt concrete mixture. It is worth mentioning that even if the fillers are in line with the requirement stated in the standard, their use in bituminous mixture needs to be further and properly investigated. Only after a fulfilling investigation, it is possible to assess how the bio fillers interacts with the other components to ensure that the final material meets the performance criteria specified by technical standards.

Globally the following conclusions can be drawn based on the experimental results obtained:

- The biofillers analysed has geometric characteristics in line with the European standard EN 13043 in terms of maximum size dimensions. Since the size and shape affect the mechanical properties of bituminous mixture, a particle shape investigation through SEM could be relevant in order to examine the influence of the particles in the final behaviour of the asphalt concrete (Sangiorgi et al., 2014).
- The results of the water content of the three materials are comparable with the traditional filler. The water content is strictly associated with the exposure to a humid environment mainly during storage. In asphalt plant production it is recommend to have humidity under control in order to ensure a correct mixing process.
- The Rigden voids are in line with the lower limit indicated in the European standard, and lower if compared to the limestone filler. Thus, in the mix design of a hypothetic asphalt concrete, the optimum bitumen content is supposed to be properly studied in case of total substitution on limestone filler with the tested biofillers, considering their absorption properties.
- From the Delta Ring and Ball test, a slightly difference is detected between the results of the three biofillers. Therefore, the innovative materials can be classified in the same range, and a similar rheological behaviour is expected.
- For every biofiller the water solubility is higher than the traditional limestone filler, but still within the limit suggested in the EN 13043. These results are certainly connected to their chemical composition that even if composed manly by $CaCO_3$, is constituted of small traces of different materials.
- High quantities of calcium carbonate for each bio fillers have been detected, as expected. Considering the renown affinity of $CaCO_3$ with bitumen, these results are promising for future applications of the experimental biofillers within bituminous mixtures. However, to obtain more specific results in terms of chemical characteristics a deeper investigation with Energy-dispersive X-ray Spectroscopy is suggested.

The laboratory investigation has highlighted that the physical, chemical and mechanical properties of the bio fillers are comparable to those verified for the limestone one. The tests suggest that the nature of the bio fillers slightly influences their characteristics.

4.1. *Prospects for future research*

Future studies will identify which properties of the filler directly impact the performance of the final asphalt mixture. In terms of mechanical characterization, the next phase in the research is planned to focus on the characterization of bituminous mastics and asphalt concretes produced with the tested biofillers in total or partial substitution of the limestone filler.

Since this study has demonstrated that transformed shells can be effectively used as fillers for bituminous mixtures, the next step will be to analyse their use through an integration of Life Cycle Assessment (LCA) and Life Cycle Cost (LCC) analysis to assess environmental impacts and potential economic benefits.

The LCA analysis will allow comparing the environmental impacts of using shells versus traditional limestone fillers. Furthermore, research conducted in Brazil suggests a maximum distance of 323 km between the shell collection site and the processing plant to achieve overall environmental benefits (Alonso et al., 2021). Based on this finding, a study on a real-scale application of the reuse, taking into account the distances between mollusc farms and a processing facility will be implemented.

Additionally, a critical point during the LCA analysis could be related to the use of high temperatures to treat the shells. Future studies could evaluate the effectiveness of shell fillers without preliminary thermal treatment, analysing their technical and environmental performances.

Integrating LCA and LCC analyses will provide a comprehensive view of shell utilization as fillers, allowing for an in-depth assessment of advantages and disadvantages from both environmental and economic perspectives, aiming to establish a solid basis for the reuse of raw materials in a circular economy framework.

CRediT authorship contribution statement

L. Caroscio: Writing – original draft, Investigation, Formal analysis.

B. De Pascale: Writing – original draft, Investigation, Formal analysis, Data curation. **P. Tataranni:** Writing – review & editing, Supervision, Methodology, Data curation. **C. Chiavetta:** Supervision, Resources, Methodology, Conceptualization. **C. Lantieri:** Supervision. **A. Bonoli:** Supervision, Resources.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

No data was used for the research described in the article.

References

Alharthai, M., Lu, Q., Elnihum, A., Elmagarhe, A., 2021. Laboratory evaluation of the use of Florida washed shell in open-graded asphalt mixtures. Materials 14. https://doi.org/10.3390/ma14227060.

Alonso, A.A., Álvarez-Salgado, X.A., Antelo, L.T., 2021. Assessing the impact of bivalve aquaculture on the carbon circular economy. J. Clean. Prod. 279, 123873 https://doi.org/10.1016/J.JCLEPRO.2020.123873.

Antunes, V., Freire, A.C., Quaresma, L., Micaelo, R., 2017. Evaluation of waste materials as alternative sources of filler in asphalt mixtures. Mater. Struct. Materiaux et Constructions 50. https://doi.org/10.1617/s11527-017-1126-3.

Arabani, M., Babamohammadi, S., Azarhoosh, A.R., 2015. Experimental investigation of seashells used as filler in hot mix asphalt. Int. J. Pavement Eng. 16, 502–509. https://doi.org/10.1080/10298436.2014.943132.

Ayunaning, K., Avisha, G.P., Juliati, K., Kunci, K., 2022. Use of Shells Waste Filler in Pavement Layer Using Lapen (Macadam Penetration Layer) Penggunaan Filler Limbah Kulit Kerang Pada Lapisan Perkerasan Dengan Menggunakan Lapen. Lapis Penetrasi Makadam.

Azra, M.N., Okomoda, V.T., Tabatabaei, M., Hassan, M., Ikhwanuddin, M., 2021. The contributions of shellfish aquaculture to global food security: assessing its characteristics from a future food perspective. Front. Mar. Sci. 8 https://doi.org/10.3389/fmars.2021.654897.

Bamigboye, G.O., Nworgu, A.T., Odetoyan, A.O., Kareem, M., Enabulele, D.O., Bassey, D. E., 2021. Sustainable use of seashells as binder in concrete production: prospect and challenges. J. Build. Eng. 34, 101864 https://doi.org/10.1016/J.JOBE.2020.101864.

BSI Standards Publication Aggregates for Bituminous Mixtures and Surface Treatments for Roads, Airfields and Other Trafficked Areas, 2013.

Cala, A., Leone, R., Saeli, M., 2023. Bivalve Mollusks Shells Valorisation And Recycling: market Potentiality and Novel Building Products.

Çatli, A.U., Bozkurt, M., Küçkyilmaz, K., Çinar, M., Bintas, E., Çöven, F., Atik, H., 2012. Performance and egg quality of aged laying hens fed diets supplemented with meat and bone meal or oyster shell meal. S. Afr. J. Anim. Sci. 42, 74–82. https://doi.org/10.4314/sajas.v42i1.9.

Çevrim, K., Iskender, E., 2022. Investigation of the usability of aggregates produced from sea shell in SMA pavements with different addition methods. Road Mater. Pavement Des. https://doi.org/10.1080/14680629.2022.2111337.

Choudhary, J., Kumar, B., Gupta, A., 2021. Potential utilization of construction wastes in asphalt pavements as fillers using ranking framework. Construct. Build. Mater. 277 https://doi.org/10.1016/j.conbuildmat.2021.122262.

Choudhary, J., Sukhija, M., Gupta, A., 2022. A comparative analysis of engineering and economical suitability of bituminous mastics containing waste fillers. Case Stud. Constr. Mater. 17, e01640 https://doi.org/10.1016/J.CSCM.2022.E01640.

Corbau, C., Nardin, W., Vaccaro, C., Vona, I., Simeoni, U., 2023. Experimental design and field deployment of an artificial bio-reef produced by mollusk shell recycling. Mar. Environ. Res. 183 https://doi.org/10.1016/j.marenvres.2022.105833.

Currey, J.D., 1999. The design of mineralised hard tissues for their mechanical functions. J. Exp. Biol. 202, 3285–3294. https://doi.org/10.1242/jeb.202.23.3285.

De Pascale, B., Tataranni, P., Lantieri, C., Bonoli, A., Sangiorgi, C., 2023. Innovative light-coloured porous asphalt for low-impact pavements: a laboratory investigation. Construct. Build. Mater. 368, 130482 https://doi.org/10.1016/J.CONBUILDMAT.2023.130482.

Diokhane, A., Sow, D., Khalil Cisse, I., 2022. Valorization of materials from road wastes: case of the seashell aggregate. Int. J. Adv. Res. 10, 546–551. https://doi.org/10.21474/IJAR01/15071.

Directive 2008/98/EC of the European Parliament and of the Council of 19 November 2008 on Waste and Repealing Certain Directives (Text with EEA Relevance), 2008.

Eziefula, U.G., Ezeh, J.C., Eziefula, B.I., 2018. Properties of seashell aggregate concrete: a review. Construct. Build. Mater. 192, 287–300. https://doi.org/10.1016/j.conbuildmat.2018.10.096.

Fan, G., Liu, H., Liu, C., Xue, Y., Ju, Z., Ding, S., Zhang, Y., Li, Y., 2022. Analysis of the influence of waste seashell as modified materials on asphalt pavement performance. Materials 15. https://doi.org/10.3390/ma15196788.

Guo, Y., Wang, X., Ji, G., Zhang, Y., Su, H., Luo, Y., 2021. Effect of recycled shell waste as a modifier on the high-and low-temperature rheological properties of asphalt. Sustainability 13. https://doi.org/10.3390/su131810271.

Jović, M., Mandić, M., Šljivić-Ivanović, M., Smičiklas, I., 2019. Recent trends in application of shell waste from mariculture. Studia Marina 47–62. https://doi.org/10.5281/zenodo.3274471 n.d.

Jyothi, M., Patel, S., Monikkah, A., Professor, A., 2018. Partial replacement of coarse aggregates using cockle seashells in flexible pavement. www.ijcrt.org.

Lagos-Varas, M., Movilla-Quesada, D., Raposeiras, A.C., Arenas, J.P., Calzada-Pérez, M. A., Vega-Zamanillo, A., Lastra-González, P., 2020. Influence of limestone filler on the rheological properties of bituminous mastics through susceptibility master curves. Construct. Build. Mater. 231, 117126 https://doi.org/10.1016/J.CONBUILDMAT.2019.117126.

Lv, S., Hu, L., Xia, C., Cabrera, M.B., Guo, Y., Liu, C., You, L., 2021. Recycling fish scale powder in improving the performance of asphalt: a sustainable utilization of fish scale waste in asphalt. J. Clean. Prod. 288, 125682 https://doi.org/10.1016/J.JCLEPRO.2020.125682.

Martínez-García, C., González-Fonteboa, B., Martínez-Abella, F., Carro- López, D., 2017. Performance of mussel shell as aggregate in plain concrete. Construct. Build. Mater. 139, 570–583. https://doi.org/10.1016/J.CONBUILDMAT.2016.09.091.

Mathews, H., Uddin, M.J., Hargis, C.W., Smith, K.J., 2023. First-year performance of the pervious Oyster Shell Habitat (POSH) along two energetic Shorelines in Northeast Florida. Sustainability 15. https://doi.org/10.3390/su15097028.

Modupe, A.E., Fadugba, O.G., Busari, A.A., Adeboje, A.O., Aladegboye, O.J., Alejolowo, O.O., Chukwuma, C.G., 2021. Sustainability assessment of the engineering properties of asphalt concrete incorporating pulverized snail shell ash as partial replacement for filler. In: IOP Conf Ser Earth Environ Sci. IOP Publishing Ltd. https://doi.org/10.1088/1755-1315/665/1/012057.

Morris, J.P., Backeljau, T., Chapelle, G., 2019. Shells from aquaculture: a valuable biomaterial, not a nuisance waste product. Rev. Aquacult. 11, 42–57. https://doi.org/10.1111/raq.12225.

Moustafa, H., Youssef, A.M., Duquesne, S., Darwish, N.A., 2017. Characterization of bio-filler derived from seashell wastes and its effect on the mechanical, thermal, and flame retardant properties of ABS composites. Polym. Compos. 38, 2788–2797. https://doi.org/10.1002/pc.23878.

Nciri, N., Shin, T., Lee, H., Cho, N., 2018. Potential of waste oyster shells as a novel biofiller for hot-mix asphalt. Appl. Sci. 8 https://doi.org/10.3390/app8030415.

Oso, A.O., Idowu, A.A., Niameh, O.T., 2011. Growth response, nutrient and mineral retention, bone mineralisation and walking ability of broiler chickens fed with dietary inclusion of various unconventional mineral sources. J. Anim. Physiol. Anim. Nutr. 95, 461–467. https://doi.org/10.1111/j.1439-0396.2010.01073.x.

Osorio-López, C., Seco-Reigosa, N., Garrido-Rodríguez, B., Cutillas-Barreiro, L., Arias-Estévez, M., Fernández-Sanjurjo, M.J., Álvarez-Rodríguez, E., Núñez-Delgado, A., 2014. As(V) adsorption on forest and vineyard soils and pyritic material with or without mussel shell: kinetics and fractionation. J. Taiwan Inst. Chem. Eng. 45, 1007–1014. https://doi.org/10.1016/j.jtice.2013.10.001.

Regulation (EU) No 1069/2009 of the European Parliament and of the Council Laying Down Health Rules as Regards Animal By-Products and Derived Products Not Intended for Human Consumption and Repealing Regulation (EC) No 1774/2002 (Animal By-Products Regulation), 2009.

Regulation (EU) No 305/2011 of the European Parliament and of the Council of 9 March 2011 Laying Down Harmonised Conditions for the Marketing of Construction Products and Repealing Council Directive 89/106/EEC (Text with EEA Relevance), 2011.

Ruiz, G., Chávez, F., Santamaría, S., Araujo, W., Timaná, J., Schmitt, R., 2020. Laboratory evaluation of seashells used as fine aggregate in hot mix asphalt. Int. J. Pavement Eng. 21, 620–628. https://doi.org/10.1080/10298436.2018.1502435.

Russo, F., Oreto, C., Veropalumbo, R., 2022. Promoting resource conservation in road flexible pavement using jet grouting and plastic waste as filler. Resour. Conserv. Recycl. 187, 106633 https://doi.org/10.1016/J.RESCONREC.2022.106633.

Sakr, M., Nazir, A., Azzam, W., Osama, N., Sakr, M.A., Nazir, A.K., Azzam, W.R., Ali, N. O., 2022. Improving the soft clay using seashell and eggshell for road construction. https://www.researchgate.net/publication/362183306.

Sangiorgi, C., Tataranni, P., Simone, A., Vignali, V., Lantieri, C., Dondi, G., 2014. Waste bleaching clays as fillers in hot bituminous mixtures. Construct. Build. Mater. 73, 320–325. https://doi.org/10.1016/j.conbuildmat.2014.09.076.

Sangiorgi, C., Tataranni, P., Simone, A., Vignali, V., Lantieri, C., Dondi, G., 2016. Assessment of waste bleaching clay as alternative filler for the production of porous asphalts. Construct. Build. Mater. 109, 1–7. https://doi.org/10.1016/j.conbuildmat.2016.01.052.

Sangiorgi, C., Tataranni, P., Mazzotta, F., Simone, A., Vignali, V., Lantieri, C., 2017. Alternative fillers for the production of bituminous mixtures: a screening investigation on waste powders. Coatings 7. https://doi.org/10.3390/coatings7060076.

Seesanong, S., Wongchompoo, Y., Boonchom, B., Sronsri, C., Laohavisuti, N., Chaiseeda, K., Boonmee, W., 2022. Economical and environmentally friendly track of biowaste recycling of scallop shells to calcium lactate. ACS Omega 7, 14756–14764. https://doi.org/10.1021/acsomega.2c00112.

Shetty, P.P., Rao, A.U., Pai, B.H.V., Kamath, M.V., 2023. Performance of high-strength concrete with the effects of seashell powder as binder replacement and waste glass powder as fine aggregate. J. Compos. Sci. 7, 92. https://doi.org/10.3390/jcs7030092.

Sholichin, I., Ayunaning, K., 2019. Utilizing seashells waste as filler on concrete asphalt mixture. Int. J. Sci. Eng. Invest. 8. www.IJSEI.com.

Summa, D., Lanzoni, M., Castaldelli, G., Fano, E.A., Tamburini, E., 2022. Trends and opportunities of bivalve shells' waste valorization in a prospect of circular blue bioeconomy. Resources 11. https://doi.org/10.3390/resources11050048.

Targino Bezerra, Ulisses, Almeida, Fábio Luiz P., Silva, Lucineide Balbino, Barbosa, Normando Perazzo, Passos, Tibério A., Cavalcante, Danielle G.L., 2011. Production of filler aggregate from waste of bivalves molluscs shells. J. Civ. Eng. Architect. 5 https://doi.org/10.17265/1934-7359/2011.04.008.

Tayeh, B.A., Hasaniyah, M.W., Zeyad, A.M., Yusuf, M.O., 2019. Properties of concrete containing recycled seashells as cement partial replacement: a review. J. Clean. Prod. 237, 117723 https://doi.org/10.1016/J.JCLEPRO.2019.117723.

The Eu fish market, Marit. Aff. Fisheries, (n.d.). https://doi.org/10.2771/38507.

The state of World fisheries and aquaculture 2022. FAO, 2022. https://doi.org/10.4060/cc0461en.

Tiwari, N., Rondinella, F., Satyam, N., Baldo, N., 2023. Alternative fillers in asphalt concrete mixtures: laboratory investigation and machine learning modeling towards mechanical performance prediction. Materials 16. https://doi.org/10.3390/ma16020807.

Topini, D., Toraldo, E., Andena, L., Mariani, E., 2018. Use of recycled fillers in bituminous mixtures for road pavements. Construct. Build. Mater. 159, 189–197. https://doi.org/10.1016/J.CONBUILDMAT.2017.10.105.

Wu, J., Liu, Q., Wu, S., 2022. Bionic synthesis of mussel-like adhesive L-DMA and its effects on asphalt properties. Materials 15. https://doi.org/10.3390/ma15155351.

Yoris-Nobile, A.I., Slebi-Acevedo, C.J., Lizasoain-Arteaga, E., Indacoechea-Vega, I., Blanco-Fernandez, E., Castro-Fresno, D., Alonso-Estebanez, A., Alonso-Cañon, S., Real-Gutierrez, C., Boukhelf, F., Boutouil, M., Sebaibi, N., Hall, A., Greenhill, S., Herbert, R., Stafford, R., Reis, B., van der Linden, P., Gómez, O.B., Meyer, H.S., Franco, J.N., Almada, E., Borges, M.T., Sousa-Pinto, I., Tuaty-Guerra, M., Lobo-Arteaga, J., 2023. Artificial reefs built by 3D printing: systematisation in the design, material selection and fabrication. Construct. Build. Mater. 362 https://doi.org/10.1016/j.conbuildmat.2022.129766.