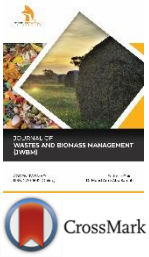
ZIBELINE INTERNATIONAL™ PUBLISHING
ISSN: 2710-6012 (Online)
CODEN: JWBMAO

Journal of Wastes and Biomass Management (JWBM)

DOI: http://doi.org/10.26480/jwbm.02.2025.47.53



RESEARCH ARTICLE

USE OF BIOGENIC CALCIUM CARBONATE FROM MOLLUSK SHELL WASTE FOR THE PRODUCTION OF GEOPOLYMERS

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ARTICLE DETAILS

Article History:

Received 23 May 2025
Revised 18 June 2025
Accepted 30 July 2025
Available online 12 August 2025

ABSTRACT

The increasing production of sea-derived waste connected with the catering industry could be used as a secondary source of raw materials, thus reducing the burden of its accumulation, which accounts for millions of tons per year, thus offering a circular economy approach. This study focuses on the possibility of retrieving calcium carbonate from seashells waste, including oysters, mussels and clams bivalve mollusks from the Adriatic Sea farming to use them into geopolymer matrices. Seashells were analyzed by means of X-ray Powder Diffraction (XRPD), and Raman spectroscopy, highlighting compositional differences, spanning from almost pure calcite for oysters and pure aragonite for clams with mussels shells having an intermediate composition (76% calcite, 24% aragonite). Up to 50 wt.% seashells were inserted into metakaolin-based geopolymer matrices as aggregate, using grain sizes from 80 microns to 2 millimeters. Mechanical tests showed that oyster-based samples have the highest flexural resistance (8 ± 0.4 MPa) at 28 days curing, whilst clam-based samples display the highest compressive resistance (60.2 ± 2.9 MPa) in the same conditions. Furthermore, physical properties, including density and open porosity of the geopolymers, indicate that clam-based samples are denser compared to oysters and mussels samples, which in turn display slightly higher porosities. The results indicate that geopolymers can incorporate high quantities of seashell waste and may be suitable for construction industry, in replacement of Portland concrete, with only limited difference in properties among the geopolymers filled by various seashell powders. This suggests even the possibility to use these waste aggregates without preliminary separation by species.

KEYWORDS

Geopolymers; seashell powder; biogenic calcium carbonate

1. INTRODUCTION

In recent years, disposing of an increasingly larger amount of sea-derived waste in a way to offer potential for their upcycling and economic value, has become an issue of interest in the field of materials (Santulli et al., 2023). In the specific case of mollusk shells, their steadily growing availability is also related to the increasing diffusion of aquaculture as opposed to fishing, with the former ensuring rather constant trends in the product supply and then eventually the need for waste disposal (Dauda et al., 2019). To ensure some real sustainability to this process, it is essential to offer processing to large amounts of this secondary raw material, which spends relatively little energy for the purpose (Morris et al., 2019). The use of mussel shells as a source of calcium carbonate through a calcination phase has been documented, though its sustainability might be limited, since this requires an increase of the temperature up to 600 °C to destroy the most resistant organic matter, without inducing further decomposition of calcium carbonate into calcium oxide and carbon dioxide (Piras et al., 2024). Another possibility explored is the production of nanosized calcium carbonate powders from mollusk shells using high-energy ball-milling in bleaching solutions (Lu et al., 2015). However, the purity of calcium carbonate in seashells can be generally deemed sufficient for further uses in materials, even with a bare protein removal and without any calcination process (Bellei et al., 2023). This opportunity arises as part of a broader context, where biogenic calcium carbonate represents a substitute material that is now widely used, given its considerable availability in food production and post-consumer systems (Barros et al., 2009). For example, an application that is gradually becoming of interest would suggest using it as a hardener in the field of polymer composites, where sustainable additives are actively sought for (Nurazzi et al., 2024). It is no surprise that this rather controllable and traceable waste stream from aquaculture raised the recent interest of the construction industry (Peceño et al., 2022), where also the presence of trace elements in seashells may not be detrimental to the performance of the construction materials developed by their use (Hart, 2020). Nonetheless, some concerns were raised over the durability of mollusk shell powder when inserted in concrete as a substitute for aggregates (Naik et al., 2024).

The interest in developing geopolymers, which are now perceived as a possible more sustainable replacement for ordinary Portland cement (OPC), is mainly linked to the possibility of minimizing the use of fossil fuels and the energy required for the production of construction materials with properties comparable to OPC (Shehata et al., 2021). The geopolymerization process, proved over the years to be also a suitable method for the durable incorporation of various waste in the material, therefore resulting in a beneficial effect on the preservation of mineral resources (de Oliveira et al., 2022). Amongst the waste types that have demonstrated their potential when being immobilized in geopolymers are fly ash, also mixed with slag, red mud, silica rich-biomass and generally ashes, even if containing heavy metals, whose content need to be controlled (Abdullah et al., 2011; John et al., 2021; Kumar et al., 2021; Liu et al., 2022; Arunachalam et al., 2022). In this sense, waste obtained from the metallurgy of non-ferrous metals, such as aluminum or zinc, proved typically suitable for introduction into geopolymers (Singh and Singh,

Quick Response Code	Access this article online	
	Website: www.jwbm.com.my	DOI: 10.26480/jwbm.02.2025.47.53

Cite the Article: Mattia Latini, Sara Mattiello, Roberto Ercoli, Paola Stabile, Carlo Santulli, Eleonora Paris (2025) Use Of Biogenic Calcium Carbonate From Mollusk Shell Waste For The Production Of Geopolymers. *Journal of Wastes and Biomass Management, 7(2):* 47-53.

2019). Even waste plastic found application in geopolymers, defined as "plastic supplemented", including both thermoplastics and thermosetting materials, which indicates their adaptability also to host mixed refuse (Panda et al., 2024).

Structural modifications can also be obtained by the use of waste-filled geopolymers, such as it is in the case of geopolymer foams obtained by a 1:1 wt.% ratio of metakaolin/silica matrix, filled with chopped carbon fibers and secondary by-products of the aluminum industry (Ercoli et al., 2022). The presence of mixed waste categories in geopolymers does not in itself represent a limitation to their functionality, as demonstrated with the introduction of 60% wt. CDW (Construction and Demolition Waste), using metakaolin as the precursor and potassium silicate as an alkaline reagent (Volpintesta et al., 2022). This suggested that even fine fractions can be employed to be added to geopolymers.

The use of mollusk shell fragments to be used as aggregate in the production of geopolymers does appear therefore a recycling strategy, which has a substantial potential for success. In the specific case of this work, the waste of three mollusk species is proposed for use, namely oysters (Ostrea edulis), mussels (Mytilus galloprovincialis), and clams (Ruditapes decussatus), all originating from the central area of the Adriatic Sea (Italy). The respective properties of the various fillers need to be compared though in their introduction into geopolymers, to assess the performance of the final materials.

2. Materials And Methods

2.1 Materials

Three types of mollusk seashell waste from oysters, mussels and clams were preliminary washed in tap water and dried at 60°C for 24 hours in a muffle furnace to eliminate the organic residues. The seashells were then ground in a laboratory jaw-crusher and sorted by using mesh-size sieves, from a dimension of 2 millimeters down to 80 micrometers. To prepare geopolymer binders, metakaolin (as the precursor, 51 wt.%) and potassium silicate (as the activator, 49 wt.%) were mixed with 20 ml water per approximately 500 g of geopolymer, and then the three types of seashells were added to the mix, using in all cases the same quantity (50 wt.%). The procedure adopted in the lab for the geopolymers' preparation is reported in more detail in Volpintesta et al., 2023 Four types of geopolymer (GP) samples were produced, three including the same quantity of oyster, mussel and clam powder, respectively, and a fourth control type without any filler, for control purposes.

2.2 Methods

Mineralogical, microchemical, and physical-mechanical analyses were carried out to characterize the seashells and the geopolymer samples produced. The seashell fine powders, obtained by mechanical grinding in an agate mortar, were analyzed by X-ray Powder Diffraction (XRPD) using a Philips apparatus (model PW1830), using Cu Kα wavelength 1.5406 Å, working at 40 kV and 25 mA, 2theta range 2-70°, step scan 0.02°. The Reference Intensity Ratio (RIR) method was used for a quantitative analysis of the mineral species and to evaluate the sample degree of crystallinity.

Raman spectroscopy (Horiba iHR320 Spectrometer), with He-Ne gas lasers (17mW) and solid-state lasers (50mW) coupled with an Olympus BX41 microscope, operated using a 532 nm wavelength laser beam (50mW), was used to determine the presence of proteinaceous remnants and specific phases, such as nacre in the seashell.

After pouring the mixes into the molds, samples of different dimensions were obtained, to be used for the measurement of water absorption and mechanical properties. Preliminary optical microscopy and Scanning Electron Microscopy (SEM) analyses were used to verify the texture of the samples and to investigate the relationships between the aggregate and the binder. The microchemical composition was determined by using a FE-SEM Sigma300 Zeiss operating at 20 KeV potential, with a 1.23 µm/pixel resolution.

Water absorption of geopolymers was measured following the ASTM C1585-13 standard using three cubic samples of 40 mm side produced for each sample type. The samples were pre-dried at 105°C for 24 hours to obtain the initial dry weight (Wdry), then they were immersed in demineralized water for further 48 hours to establish the final (water-saturated) weight (Wsat) according to the standard prescriptions. Density measurements were carried out by an Anton Paar N-pycnometer, (Ultrapyc 5000), operating with a pressure of nitrogen gas of 1.31 bars, at 20°C. Porosity was calculated from the data obtained in the water absorption test, having the subtracted suspended weight of the samples.

For the purpose of mechanical testing, after pouring the mixes into the molds, a total of 20 prismatic samples of dimensions 160x40x40 mm were produced. The samples were covered with a plastic sheet and stored at ambient temperature for curing until testing. All samples were tested according to EN 196-1 standard at 7 and 28 days of curing. Three-point bending method and uniaxial compression method have been used by means of a Uniframe 250 Electromechanical Universal Tester (70-T2502) with a 100 kN load cell for both flexural and compressive strength evaluation of the samples. Both tests were performed in load control mode, using a load rate of 2400 (N/s), with a distance equal to 100 mm between the supports.

3. Results

The three seashell types appear macroscopically (Figure 1a-c) and microscopically (Figure 1d-f) with different colors, shapes and shell thicknesses. Oysters (Fig. 1a) show a more complex structure with respect to the other two seashells, being composed of alternate layers of calcium carbonate and nacre, in a typical "brick and mortar" typical arrangement (Yao et al., 2014) and a relatively flat and thin shape of the fragments, with sharp edges. Mussels (Figure 1b) are characterized by a thick rigid shell characterized by multiple superposed layers of calcium carbonate added with a perlaceous internal layer made of nacre (Ren et al., 2009). As a consequence, the fragments often appear as sherds with mother-of-pearl texture. Clams' (Figure 1c) structure is similar to that of mussels, although the innermost nacre layer is not so abundant: its absence might ultimately affect hardness and toughness (Cheng et al., 2023). Compared to mussels, the fragments show a less evident mother-of-pearl texture and mostly rounded edges. polymers; seashell powder; biogenic calcium carbonate

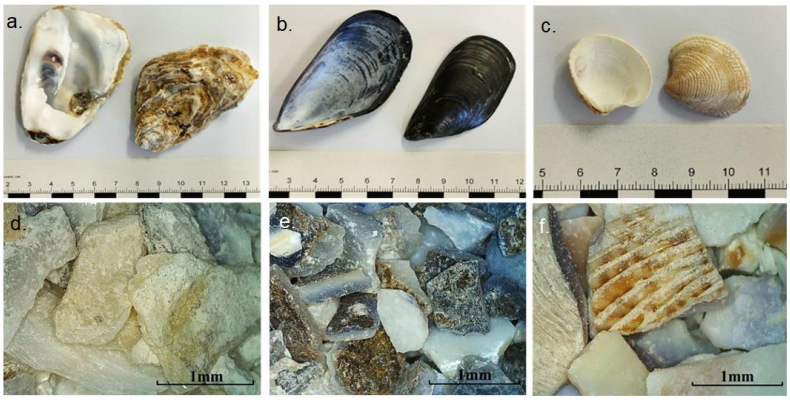

Figure 1: Optical microscope images. a-c Shells: a: Oyster shells; b: Mussel shells; c: Clam shells; d-f: Fragments: d. Oyster shell fragments; e: Mussel shell fragments; f. Clam shell fragments

X-ray diffraction data (Table 1 and Figure 2) show that, among the crystalline phases composing the shells, calcium carbonate ($CaCO_3$) polymorphs (calcite and aragonite) are both present, although in varying amounts in the three species. Calcite is highly predominant in oysters (98.5%), whereas clams are completely composed of aragonite, with mussels being intermediate between the two, but still containing prevalent amounts of calcite (75.7 wt.%) over aragonite. Quartz and halite are also present in trace amounts in the oyster shells, possibly as grains trapped in between layers during shells' growth. The amorphous content of the shells varies from the highest value of 52.4 wt.% in oysters to the lowest (39.5 wt.%) in clams. The presence of organic phases is also reported in Table 1, namely for oysters and mussels, as will be indicated further down from RAMAN results.

Table 1: Species examined and their characteristics				
Species	**Calcite (wt. %)**	**Aragonite (wt. %)**	**Amorphous fraction (wt.%)**	**Organic phases***
Oysters (*Ostrea edulis*)	98.5	1.5	52.4	Nacre, carbohydrates
Mussels (*Mytilus galloprovincialis*)	75.7	24.3	46.5	Nacre
Clams (*Ruditapes decussatus*)	n.d.	100	39.5	n.d.

Note: mineralogical data obtained by XRPD; *organic phases as determined by RAMAN; n.d. = not detected or below detection limits

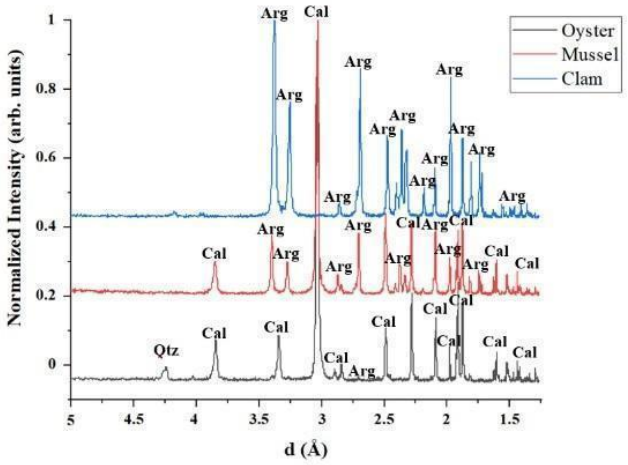

Figure 2: X-ray diffraction spectra of the different seashell samples

The aspect of geopolymer samples surface obtained by incorporation of the shell aggregates is reported in Figure 3. The optical images show the fragment homogeneously dispersed in the geopolymer matrix, with no preferential orientation and with an apparent good contact between the aggregate and binder in all samples. Some quasi-circular porosity on the sample surfaces is evident, mostly due to air bubbles incorporation during blending.

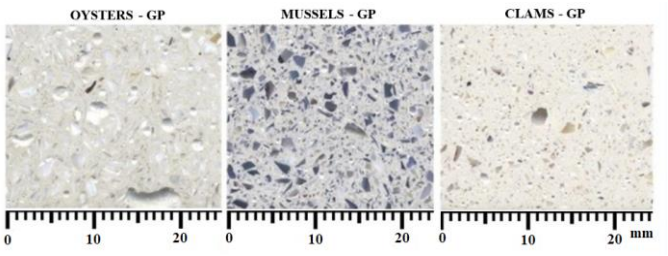

Figure 3: Surface of the geopolymer samples containing the three different seashell waste

SEM images (Figure 4a) show in more detail the effective good contact between grains and binder even at a much smaller scale, demonstrating also the lack of fractures or discontinuities in the matrix and evidencing the absence of reaction rims or bubbles incorporation between fragments and matrix. The EDS elemental maps (Figure 4 b-e) of all samples show the chemical composition of aggregate and matrix: the presence of Ca (red) is referred to seashell $CaCO_3$-rich grains, whilst Al, Si, and K are related to the geopolymer matrix composed of metakaolin and potassium silicate, The matrix appears to be chemically homogeneous and well distributed in the sample, without crystallization of new phases.

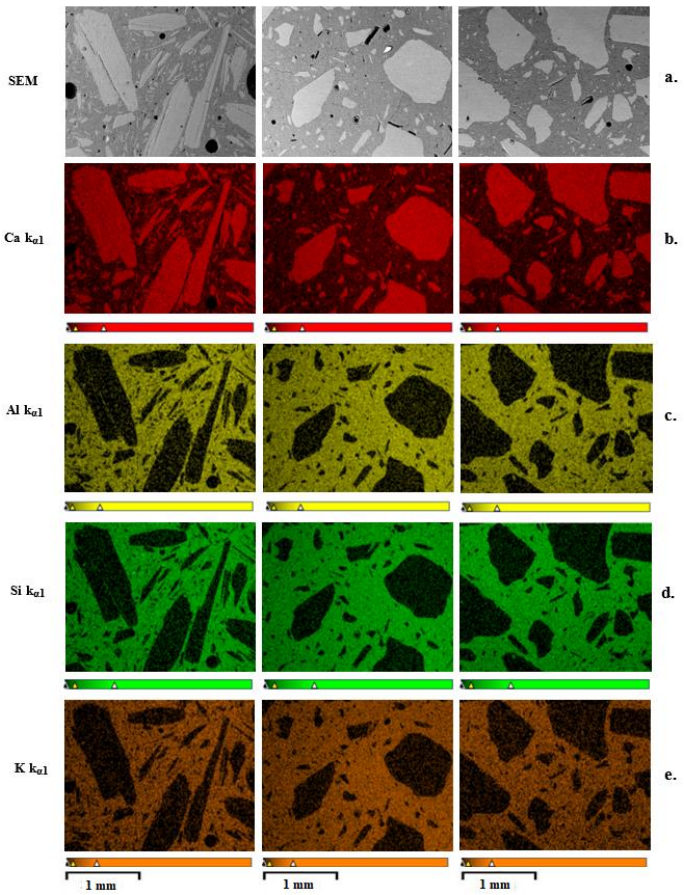

Figure 4: SEM images and microchemical data for Oysters-GP sample (left column), Mussels-GP sample (center), Clams-GP sample (right column). (a) SEM backscattered images; (b, c, d, e) EDS elemental maps of GP samples: (b) calcium; (c) aluminum; (d) silicon; (e) potassium

The Raman spectra show (Figure 5) the peak relative to metakaolin at 150-148 cm^{-1} as overlapped with the peak related to aragonite for all the geopolymers with seashell fragments (Mahrous et al., 2024; Parker et al., 2010). Other peaks related to aragonite and calcite are reported in Table 2. In geopolymers including mussels (Figure 5b) and oysters (Figure 5c), the organic fraction is more visible than in the case of the geopolymer with clams (Figure 5a). Furthermore, the band in position ~ 1090 cm^{-1} was assigned to the Si-O-Si symmetric vibration of the amorphous silica related to metakaolin. [De Leeuw and Parker 1998).

The peak in the band between 1484 and 1469 cm^{-1} is visible in all three samples and is attributed to the organic functional group; this band is in general assigned to C=O or to C=C bond vibrations, in agreement with the presence of biomolecules (Ubaldini et al., 2024). The peak at 998 cm^{-1} attributed to phenylalanine contained in collagen (Sikes et al., 1998) is only present in the geopolymers with mussels and oysters, and finally only for the geopolymer with mussels at 1602 cm^{-1} amide I band, related to proteins (Talari et al., 2015) is visible. All the data obtained appears in line with what was detected in the X-ray diffraction spectra.

Table 2: Peak assignment of Raman spectra for GP with the three types of seashells and the literature used.

Wavenumbers (cm^{-1})			Assignment	References
Oysters-GP	Mussels-GP	Clams-GP		
148	146	150	MK or aragonite	Mahrous et al., 2024; Parker et al., 2010
		208	aragonite	Parker et al., 2010
284	285		calcite	Parker et al., 2010
701	704	701	aragonite	Parker et al., 2010
1006	998		phenylalanine	Sikes et al., 1998
		1086	aragonite	Talari et al., 2015
1096	1097		Si-O-Si	de Leeuw & Parker, 1998
1469	1484	1497	Organic functional group	Gupta et al., 2024
	1602		Amide I	Talari et al., 2015

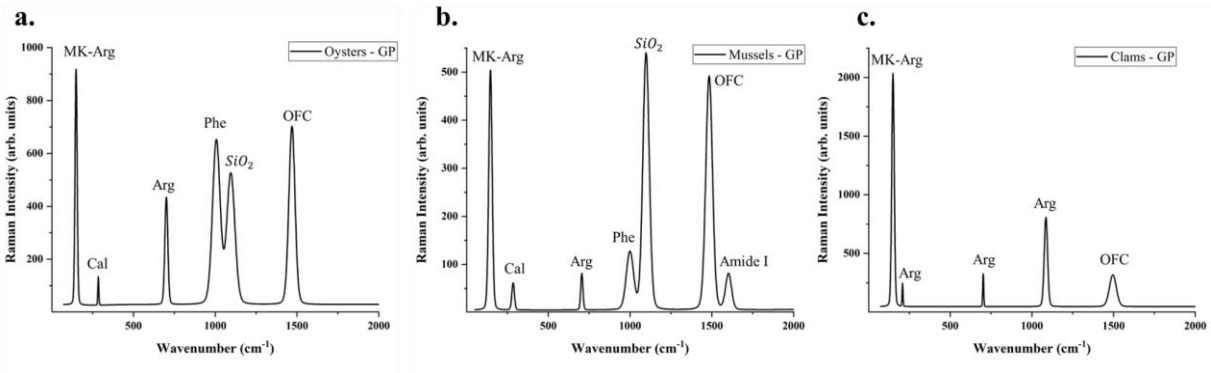

Figure 5: Raman spectra of seashell-bearing geopolymers: a) Oysters-GP; b) Mussels-GP; c) Clams-GP MK=Metakaolin, Cal = Calcite, Arg = Aragonite, OFC=organic functional group, Phe= phenylalanine, SiO^2= amorphous silica.

The physical properties of the geopolymer samples were measured in terms of density, porosity, and water absorption (Table 2). Density data (D) indicate that clam-based geopolymer is denser than the oysters- and mussels-based ones, which directly reflect their calcite and aragonite contents. In fact, since pure calcite and pure aragonite have a density of 2.71 and 2.96 g/cm^3, respectively, due to the higher coordination number of the Ca ions in the aragonite structure [32], the GP samples density increases with the aragonite content of the seashells aggregate. Porosity data and water absorption capacity show highest values for oyster-based geopolymers followed by mussels-GP and clams-GP, with only slight differences. The control-GP sample shows the lowest density and higher porosities/water absorption with respect to the seashell-based geopolymers.

Mechanical tests, in terms of compressive and flexural strength, were performed at 7 and 28 days of curing. All samples display good mechanical properties already at 7 days (over 45 MPa for compressive strength and over 6 MPa for flexural strength, respectively) and reach a further overall increase at 28 days. Regarding compressive strength, clam-based geopolymers display the highest values of resistance after 28 days, which is comparable with the values obtained for the control sample. In contrast, the highest flexural strength is obtained by oyster-based samples, which outperform the control sample.

Table 3: Physical and mechanical properties of seashells geopolymers

Sample	Density (g/cm^3)	Porosity (%)	Water absorption (%)	Compressive strength (MPa) 7 days	28 days	Flexural strength (MPa) 7 days	28 days
Oysters GP	2.47 ± 0.02	15.4 ± 0.1	15.3 ± 0.7	46.1 ± 3.6	49.9 ± 1.7	6.5 ± 0.3	8 ± 0.4
Mussels GP	2.49 ± 0.05	14.6 ± 0.8	14.6 ± 0.9	45.8 ± 1.2	46.4 ± 3.7	7.3 ± 0.1	6.8 ± 0.1
Clams GP	2.58 ± 0.03	13.7 ± 0.9	13.6 ± 1	53.5 ± 1.1	60.2 ± 2.9	6.3 ± 0.4	7.3 ± 0.4

Table 3(Cont.): Physical and mechanical properties of seashells geopolymers							
Control GP	1.80 ± 0.01	21.3 ± 3	21.3 ± 2.3	-	62.1 ± 3.6	-	5.6 ± 1.1

Figure 6 reveals that all the transverse sections of fractured geopolymer samples filled with seashell powder indicate the presence of multiple crack planes from which the breakage process develops (Figure 6 a, b, c). This suggests a limited brittleness and the ability of fillers to improve the samples' toughness. In contrast, the control-GP sample (Fig. 6d) reveals the obvious presence of propagating cracks, whose action is enhanced by the higher amount of porosity, and leads to a sudden failure.

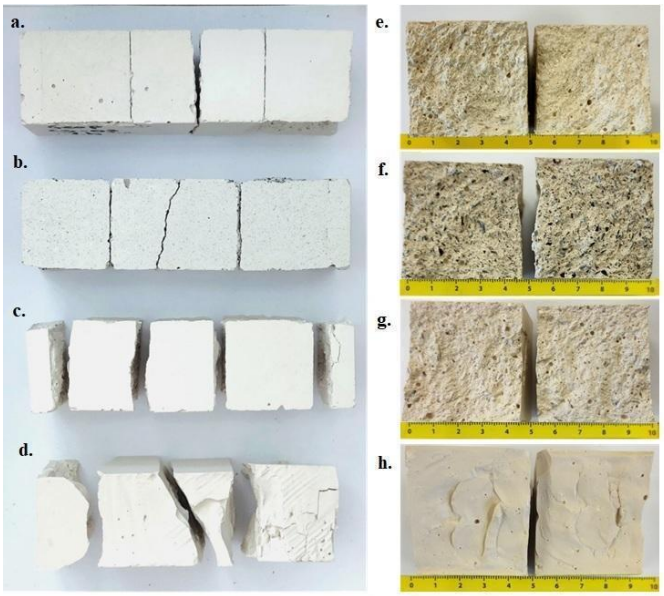

Figure 6: Fractured flexural samples: (a, e) Oysters GP sample (longitudinal and transverse); (b, f) Mussels GP sample (longitudinal and transverse); (c, g) Clams GP sample (longitudinal and transverse); (d, h) Unfilled control-GP sample (longitudinal and transverse)

4. Discussion

Observations with optical microscopy of seashell fragments highlight a platy-like morphology of oysters' shell grains with multiple superposed layers of calcium carbonate. This type of brick-and-mortar structure may favor a more ductile behaviour of the samples under load, thus explaining the high flexural resistance reached by oyster-based geopolymers (Gupta et al., 2024). The presence of nacre and organic remains in the oysters shells may also account for its lower density and higher porosity, in contrast with denser and less porous mussels and clams seashells.

The limited differences in mechanical and physical properties of the seashell-filled geopolymer samples can be correlated with the variable mineralogical characteristics of the aggregate added. The three types of seashells (oysters, mussels and clams) differ in terms of calcium carbonate polymorphs (calcite-aragonite) contents: mechanical test results indicated that the geopolymer added with oyster shell powder offered the highest value for flexural resistance after 28 days of curing. This is likely to indicate that the complex topography of oyster shells, correlated with the higher content of nacre present in these by its specific mineralization process (Sun and Bhushan, 2012), as evidenced by Raman spectroscopy analysis, can contribute to a more effective behavior also in geopolymers. The high compressive strength of clam-filled geopolymers can be explained by the purely aragonitic composition of the shell, which is the denser and tougher calcium carbonate polymorph. Furthermore, no organic matter residue has been detected in clam seashell, possibly due to a very low concentration below detection limits. This may in turn justify the higher density of the sample, thus enhancing compressive resistance. On the other hand, clamshell powder has been demonstrated to be a particularly effective filler e.g., when coupled with natural fibers in polymers (Jena and Panigrahi, 2021). In contrast, the fact that mussel-based geopolymers do not indicate a substantial increase of the properties between 7 and 28 days of the curing process might suggest that they tend to ensure a faster settling of water in the geopolymer, yet on the other hand they offer finally a slightly lower mechanical performance.

To try to compare the results obtained in this work with data obtained from literature, it is noteworthy that in the last few years, the potential introduction of biogenic ceramic fillers into geopolymers has sometimes been considered, which brought to several relevant outcomes. In particular, performed activation through sodium hydroxide solution to the partial substitution of metakaolin, which suggested that this combination was effective without material loss up to 40 wt.% of mussel or oyster shells (Monneron-Gyurits et al., 2018). More recently, pushed the substitution of metakaolin with up to 50% seashell (oysters), again activated with sodium hydroxide, a composition that offered a 28-day compression and flexural strength equal to 23.2 and 3.4 MPa, respectively (Assaad and Saba, 2023). The researchers assessed the feasibility of total substitution of geopolymer aggregates with oyster shells, which brought to a markedly higher result, namely 67 and 6.3 MPa for compression and flexural strength, respectively: this was attributed by a high degree of connected pore tortuosity and pore wall roughness in the geopolymer, enhanced by oyster shell porosity (Yanting et al., 2025).

Alongside the aforementioned results, more classical applications are continuously proposed for the introduction of seashell powder in Portland concrete, alas with lower levels of substitution to preserve the mechanical performance. In particular, as regards mussels, mechanical improvements were revealed by substitution of up to 6% cement, oysters with effective substitution of up to 15% and also for a hybrid solution, considering clams in combination with cockle shells, where the degree of replacement was limited to 4% (Stel'makh et al., 2022; Olivia et al., 2017; Ruslan et al., 2022). As a whole, though in a very cautious way, oyster shell replacement can be reported to be the most popular, also in combination with other waste, such as furnace ash, into cement, with a limited, though conceptually important, contribution to decarbonisation (Ravi et al., 2023).

Outcomes from the present study suggest that the degree of substitution with mollusk shell powders can be very high in the field of construction materials not affecting their performance and that geopolymers are particularly suitable for this objective. This is likely to become a fashionable way to reduce the environmental impact both for this type of food waste, which is normally landfilled, and to reduce resource consumption into the construction sector, as suggested (Cardoso et al., 2023). It is likely that for this purpose a "zero km" approach is selected with locally collected material, only subjected to a limited transformation process, which can be competitive against other options, such as their calcination, as proposed (Topić Popović et al., 2023).

5. Conclusions

This study shows it is possible to recycle seashell waste into geopolymer binders, with interesting possible applications for the construction industry. Promising upcycling solutions would not only avoid landfilling of sea waste but also allow the valorization of high-grade products of Ca-carbonate-rich waste.

The introduction of 50 wt.% seashells waste aggregate (either oysters, mussels, or clams powders) into geopolymer binders produces materials with mechanical performance comparable with technical materials, which have been fully characterized in this study. The results evidenced that these materials could play a role in the future of construction by positively impacting not only the recycling chain of this waste but also by offering alternatives to the concrete production chain by adopting low-CO2 emissions geopolymers binders.

The differences obtained using various seashell fillers do not impede the possible use of these in different proportions at the same time in the same geopolymer, which would further simplify the collection process, avoiding expensive, time consuming and difficult industrial separation of the seashell waste by species. It is noteworthy, though, that resistance is enhanced after 28 days of curing especially for clam-based geopolymers, thus widening the field of applications of these new materials. Further studies could involve the use of seashells with mixed proportions into a single metakaolin geopolymer sample or even increase the quantity of aggregate, given the good performance already demonstrated, to fully uncover the potential of these waste-based materials. The use of other waste materials in the mixes, to be used as precursors substituting for metakaolin, like fly ash or others, would further enhance the green characteristic of these products, allowing to progressively reduce exploitation of raw materials, while recycling waste and impacting positively on the Life Cycle Assessment of the final product.

REFERENCES

Abdullah, M. M. A., Hussin, K., Bnhussain, M., Ismail, K. N., and Ibrahim, W. M. W., 2011. Mechanism and chemical reaction of fly ash geopolymer cement—A review. International Journal of Pure and Applied Sciences and Technology, 6(1), Pp. 35–44.

Agbaje, O. B., 2024. Microscale analysis reveals nature of hydrophobic biopolymers in aragonite bivalve shells with crossed-lamellar architecture. Microchemical Journal, 204, 111010. https://doi.org/10.1016/j.microc.2024.111010

Arunachalam, S. K., Muthiah, M., Rangaswamy, K. D., Kadarkarai, A., and Arunasankar, C. G., 2022. Improving the structural performance of reinforced geopolymer concrete incorporated with hazardous heavy metal waste ash. World Journal of Engineering, 19(6), Pp. 808–821. https://doi.org/10.1108/WJE-01-2021-0055

Assaad, J. J., and Saba, M., 2023. Use of seashell and limestone fillers in metakaolin-based geopolymers for masonry mortars. Minerals, 13(2), 186. https://doi.org/10.3390/min13020186

Barros, M. C., Bello, P. M., Bao, M., and Torrado, J. J., 2009. From waste to commodity: Transforming shells into high purity calcium carbonate. Journal of Cleaner Production, 17(3), Pp. 400–407. https://doi.org/10.1016/j.jclepro.2008.08.013

Bellei, P., Torres, I., Solstad, R., and Flores-Colen, I., 2023. Potential use of oyster shell waste in the composition of construction composites: A review. Buildings, 13(6), 1546. https://doi.org/10.3390/buildings13061546

Cardoso, A. D. S., Rabbani, E. R. K., Delmiro, T. D., Mocock, J. F. B., Silva, S. P. R. D., Filippelli, G., ... and Monteiro, E. C. B., 2023. Mollusk shell waste: Alternatives for reuse in construction. International Journal of Environment and Waste Management, 31(1), Pp. 61–80. https://doi.org/10.1504/IJEWM.2023.129727

Cheng, M., Liu, M., Chang, L., Liu, Q., Wang, C., Hu, L., ... and Chen, J., 2023. Overview of structure, function and integrated utilization of marine shell. Science of the Total Environment, 870, 161950. https://doi.org/10.1016/j.scitotenv.2023.161950

Dauda, A. B., Ajadi, A., Tola-Fabunmi, A. S., and Akinwole, A. O., 2019. Waste production in aquaculture: Sources, components and managements in different culture systems. Aquaculture and Fisheries, 4(3), Pp. 81–88. https://doi.org/10.1016/j.aaf.2018.10.002

de Leeuw, N. H., and Parker, S. C., 1998. Surface structure and morphology of calcium carbonate polymorphs calcite, aragonite, and vaterite: An atomistic approach. The Journal of Physical Chemistry B, 102(16), Pp. 2914–2922. https://doi.org/10.1021/jp973210f

de Oliveira, L. B., de Azevedo, A. R., Marvila, M. T., Pereira, E. C., Fediuk, R., and Vieira, C. M. F., 2022. Durability of geopolymers with industrial waste. Case Studies in Construction Materials, 16, e00839. https://doi.org/10.1016/j.cscm.2021.e00839

Ercoli, R., Laskowska, D., Nguyen, V. V., Le, V. S., Louda, P., Łoś, P., ... and Buczkowska, K. E., 2022. Mechanical and thermal properties of geopolymer foams (GFs) doped with by-products of the secondary aluminum industry. Polymers, 14(4), 703. https://doi.org/10.3390/polym14040703

Gupta, S., Esmaeeli, H. S., and Moini, R., 2024. Tough and ductile architected nacre-like cementitious composites. Advanced Functional Materials, 34(39), 2313516. https://doi.org/10.1002/adfm.202313516

Hart, A., 2020. Mini-review of waste shell-derived materials' applications. Waste Management and Research, 38(5), Pp. 514–527. https://doi.org/10.1177/0734242X19897812

Jena, H., and Panigrahi, A., 2021. The effect of clam shell powder on kinetics of water absorption of jute epoxy composite. World Journal of Engineering, 18(5), Pp. 684–691. https://doi.org/10.1108/WJE-08-2020-0334

John, S. K., Nadir, Y., and Girija, K., 2021. Effect of source materials, additives on the mechanical properties and durability of fly ash and fly ash-slag geopolymer mortar: A review. Construction and Building Materials, 280, 122443. https://doi.org/10.1016/j.conbuildmat.2021.122443

Kumar, A., Saravanan, T. J., Bisht, K., and Kabeer, K. S. A., 2021. A review on the utilization of red mud for the production of geopolymer and alkali activated concrete. Construction and Building Materials, 302, 124170. https://doi.org/10.1016/j.conbuildmat.2021.124170

Liu, Z., Deng, P., and Zhang, Z., 2022. Application of silica-rich biomass ash solid waste in geopolymer preparation: A review. Construction and Building Materials, 356, 129142. https://doi.org/10.1016/j.conbuildmat.2022.129142

Lu, J., Lu, Z., Li, X., Xu, H., and Li, X., 2015. Recycling of shell wastes into nanosized calcium carbonate powders with different phase compositions. Journal of Cleaner Production, 92, Pp. 223–229. https://doi.org/10.1016/j.jclepro.2014.12.093

Mahrous, M. A., Abdelghany, M. A., Trindade, A. C. C., Aboukhatwa, M., Kriven, W. M., and Jasiuk, I. M., 2024. Investigating irradiation effects on metakaolin-based geopolymer. Construction and Building Materials, 437, 136837. https://doi.org/10.1016/j.conbuildmat.2024.136837

Monneron-Gyurits, M., Joussein, E., Soubrand, M., Fondanèche, P., and Rossignol, S., 2018. Valorization of mussel and oyster shells toward metakaolin-based alkaline activated material. Applied Clay Science, 162, Pp. 15–26. https://doi.org/10.1016/j.clay.2018.05.027

Morris, J. P., Backeljau, T., and Chapelle, G., 2019. Shells from aquaculture: A valuable biomaterial, not a nuisance waste product. Reviews in Aquaculture, 11(1), Pp. 42–57. https://doi.org/10.1111/raq.12225

Naik, B., Paul, S., Choudhury, S. K., Sarangi, P. P., Mustakim, S. M., Swar, S. N., ... and Jethy, B., 2024. A comprehensive study on utilization of shell wastes in construction industry. Materials Today: Proceedings, 115, Pp. 110–114. https://doi.org/10.1016/j.matpr.2023.07.048

Nurazzi, N. M., Norrahim, M. N. F., Mulla, M. H., Kamarudin, S. H., Rani, M. S. A., Rushdan, A. I., and Kuzmin, A. M., 2024. Mechanical performance of seashell-reinforced polymer composites for structural applications. In S. M. Salit, C. H. Azhari, and N. M. Nurazzi (Eds.), Polymer composites derived from animal sources (pp. 243–257). Woodhead Publishing. https://doi.org/10.1016/B978-0-443-22414-0.00013-2

Olivia, M., Oktaviani, R., and Ismeddiyanto., 2017. Properties of concrete containing ground waste cockle and clam seashells. Procedia Engineering, 171, Pp. 658–663. https://doi.org/10.1016/j.proeng.2017.01.404

Panda, S., Nanda, A., and Panigrahi, S. K., 2024. Potential utilization of waste plastic in sustainable geopolymer concrete production: A review. Journal of Environmental Management, 366, 121705. https://doi.org/10.1016/j.jenvman.2024.121705

Parker, J. E., Thompson, S. P., Lennie, A. R., Potter, J., and Tang, C. C., 2010. A study of the aragonite-calcite transformation using Raman spectroscopy, synchrotron powder diffraction and scanning electron microscopy. CrystEngComm, 12(5), Pp. 1590–1599. https://doi.org/10.1039/B921487A

Peceño, B., Bakit, J., Cortes, N., Alonso-Fariñas, B., Bonilla, E., and Leiva, C., 2022. Assessing durability properties and economic potential of shellfish aquaculture waste in the construction industry: A circular economy perspective. Sustainability, 14(14), 8383. https://doi.org/10.3390/su14148383

Piras, S., Salathia, S., Guzzini, A., Zovi, A., Jackson, S., Smirnov, A., ... and Santulli, C., 2024. Biomimetic use of food-waste sources of calcium carbonate and phosphate for sustainable materials—A review. Materials, 17(4), 843. https://doi.org/10.3390/ma17040843

Ravi, M., Murugesan, B., and Onyelowe, K. C., 2023. Performance evaluation of marine and industrial wastes in cement to envelope low carbon environment in manufacturing process. *International Journal of Low-Carbon Technologies, 18*, Pp. 986–998. https://doi.org/10.1093/ijlct/ctad082

Ren, F., Wan, X., Ma, Z., and Su, J., 2009. Study on microstructure and thermodynamics of nacre in mussel shell. Materials Chemistry and Physics, 114(1), Pp. 367–370. https://doi.org/10.1016/j.matchemphys.2008.09.036

Ruslan, H. N., Muthusamy, K., Mohsin, S. M. S., Jose, R., and Omar, R., 2022. Oyster shell waste as a concrete ingredient: A review. Materials Today: Proceedings, 48, Pp. 713–719. https://doi.org/10.1016/j.matpr.2021.02.208

Santulli, C., Fragassa, C., Pavlovic, A., and Nikolic, D., 2023. Use of sea waste to enhance sustainability in composite materials: A review. Journal of

Marine Science and Engineering, 11(4), 855. https://doi.org/10.3390/jmse11040855

Shehata, N., Sayed, E. T., and Abdelkareem, M. A., 2021. Recent progress in environmentally friendly geopolymers: A review. Science of the Total Environment, 762, 143166. https://doi.org/10.1016/j.scitotenv.2020.143166

Sikes, C. S., Wheeler, A. P., Wierzbicki, A., Dillaman, R. M., and De Luca, L., 1998. Oyster shell protein and atomic force microscopy of oyster shell folia. The Biological Bulletin, 194(3), Pp. 304–316. https://doi.org/10.2307/1543100

Stel'makh, S. A., Shcherban', E. M., Beskopylny, A. N., Mailyan, L. R., Meskhi, B., Tashpulatov, S. S., ... and Tyutina, A., 2022. Composition, technological, and microstructural aspects of concrete modified with finely ground mussel shell powder. Materials, 16(1), 82. https://doi.org/10.3390/ma16010082

Sun, J., and Bhushan, B., 2012. Hierarchical structure and mechanical properties of nacre: A review. RSC Advances, 2(20), Pp. 7617–7632. https://doi.org/10.1039/C2RA20218B

Talari, A. C. S., Movasaghi, Z., Rehman, S., and Rehman, I. U., 2015. Raman spectroscopy of biological tissues. Applied Spectroscopy Reviews, 50(1), Pp. 46–111. https://doi.org/10.1080/05704928.2014.923902

Thompson, C. M., North, E. W., Kennedy, V. S., and White, S. N., 2015. Classifying bivalve larvae using shell pigments identified by Raman spectroscopy. Analytical and Bioanalytical Chemistry, 407, Pp. 3591–3604. https://doi.org/10.1007/s00216-015-8575-8

Topić Popović, N., Lorencin, V., Strunjak-Perović, I., and Čož-Rakovac, R., 2023. Shell waste management and utilization: Mitigating organic pollution and enhancing sustainability. Applied Sciences, 13(1), 623. https://doi.org/10.3390/app13010623

Ubaldini, A., Cicconi, F., Calistri, S., Salvi, S., Telloli, C., Marghella, G., ... and Rizzo, A., 2024. Removal of organic materials from Mytilus shells and their morphological and chemical-physical characterisation. Crystals, 14(5), 464. https://doi.org/10.3390/cryst14050464

Volpintesta, F., Finocchiaro, C., Barone, G., Mazzoleni, P., and Paris, E., 2024. Compositional differences in construction and demolition wastes (CDWs) for geopolymer mortars: A comparative study using different precursors and alkaline reagents. Minerals, 14(4), 376. https://doi.org/10.3390/min14040376

Volpintesta, F., Ossoli, E., Reggiani, A., Stabile, P., Santulli, C., and Paris, E., 2023. Geopolymers-based application for the up-cycling utilization of construction and demolition waste from the 2016 central Italy earthquakes. Materials Letters, 336, 133849. https://doi.org/10.1016/j.matlet.2023.133849

Yanting, M., Bo, D., Xiaotong, Y., Da, C., and Yingdi, L., 2025. Study on mechanical properties and durability of geopolymer concrete with oyster shell aggregate. Construction and Building Materials, 472, 140926. https://doi.org/10.1016/j.conbuildmat.2025.140926

Yao, Z., Xia, M., Li, H., Chen, T., Ye, Y., and Zheng, H., 2014. Bivalve shell: Not an abundant useless waste but a functional and versatile biomaterial. Critical Reviews in Environmental Science and Technology, 44(22), Pp. 2502–2530. https://doi.org/10.1080/10643389.2013.829763