Potential Application of Marble and Crushed Mussel Shells By-products to be Used as Aggregates in Plain Concrete Mixes

James Rombi(✉), Marta Salis, Marco Olianas, Francesca Maltinti, and Mauro Coni

Department of Civil Environmental Engineering and Architecture, University of Cagliari, 09042 Cagliari, Italy
james.rombi@unica.it

Abstract. The main objective of the following paper is to verify the possible applications of marble by-products obtained from the ornamental stone industry and mussel shells deriving from aquaculture farming to be used as alternative aggregates in plain concrete mixes. Both marble and mussel shells came from Sardinia Island (Italy), very active in the commercialization of marble for ornamental purposes and mussels from aquaculture farming. Italy is the third producer in Europe of mussel and one of the leaders worldwide together with China and Turkey for the production of marble. Marble by-products were used to replace coarse natural aggregates while the crushed mussel shells partially replaced fine aggregates. The mixes were studied to obtain non-structural concrete. Various mix designs were studied, in which marble and mussel shells, in different percentages, replaced natural aggregates (sand and gravel). The research aimed to identify the characteristics of the materials through laboratory tests and to create the optimal mix design. From the test conducted it was possible to evaluate that marble and mussel shells by-products can be considered valid alternatives to natural aggregates. From the preliminary results, encouraging mechanical performances were obtained when replacing 50% of coarse aggregates fraction 8–16 mm with marble by-products and 8% of fine aggregates 0–4 mm with crushed mussels shells. The reuse of these materials is essential in a sustainable and environmentally friendly circular economy approach, which focuses on reducing waste and reusing materials within production cycles. The resulting advantages are less exploitation of natural aggregates and less production of waste.

Keywords: By-products · Marble · Mussels shells · Concrete · Circular Economy

1 Introduction

The European Union is increasingly focused on the reuse of waste materials, that must have good mechanical and physical performance to be reused in civil engineering, but also produced with low CO_2 emissions. Based on these considerations, this work evaluated the potential applications of marble and mussel shells by-products as a valid

© The Author(s), under exclusive license to Springer Nature Switzerland AG 2023
O. Gervasi et al. (Eds.): ICCSA 2023 Workshops, LNCS 14111, pp. 210–224, 2023.
https://doi.org/10.1007/978-3-031-37126-4_15

alternative replacing natural aggregates in plain concrete mixes. The marble used in the research derives from the region of Orosei located on Sardinia Island and is purely characteristic of the eastern coastal area of the island [1]. Since the 1970s, Orosei marble has been extracted from quarries located on the slopes of Monte Tuttavista. The Monte Tuttavista deposit extends for about 300 hectares and it is currently exploited for 6% of proven reserves. In the quarrying of ornamental stones, the percentage of commercial blocks produced with respect to the volume of deposit demolished averages around 30% of the total. The remaining 70% represents scrap which, in the absence of incisive valorization activities, is partially abandoned within the site or disposed of in landfills. In the landfill area, around 3000,000 m^3 have now fallen over an area of 15.5 hectares, with evident piles of debris whose height and irregularity have created a significant impact on the landscape and compromised the functionality of the site. The mining activity has generated a significant quantity of scraps of different natures and characteristics, the arrangement of which represents a problem of considerable importance for the quarrying activities, for the environment, and for the community.

Considering bivalve molluscs they represent almost 10% of the world's total fishery production, 26% of the entire volume, and 14% of the entire value of the world's total aquaculture production [2]. Global bivalve mollusc production has increased substantially in the last fifty years, going from nearly 1 million tonnes in 1950 to about 14.6 million tonnes in 2010. China is by far the leading producer of bivalve molluscs, with 10.35 million tonnes in 2010, representing 70.8% of global molluscan shellfish production and 80% of global bivalve mollusk aquaculture production. Considering only mussel shells production China, Chile, and Spain were the top three producers considering the period 2011–2020 (see Fig. 1), providing 74% of the global production. Italy in 2020 was the third producer in Europe of mussel (12%), behind Spain (47%) and France (14%) [3]. Analyzing the different regions of Italy in which mussels shells are produced (see Fig. 2), Sardinian Island is 5^{th} in terms of production after the region of Emilia-Romagna, Veneto, Puglia, and Friuli Venezia Giulia [3, 4]. The mussel shells used in this research came from the aquaculture forming in the Gulf of Oristano, positioned on the west coast of the Island of Sardinia (see Fig. 2). As it is well known, the shell is about 33% of the entire weight of mussel shell waste. Thus, it can be confirmed that the canning industry generates over 1 million tonnes worldwide. This implies a significant waste problem on a global scale. For this reason, potential applications to use such by-products are vital to limit the disposal in landfills. In this paper potential application of marble and mussel shells by-products are studied to partially replace natural aggregates in non-structural concrete.

In recent years the construction industry has become increasingly aware of the need to change towards sustainability methods, studying and introducing new materials. Few researchers have studied the use of mussel shell waste as construction material, using it as an aggregate or filler to be incorporated into concretes or mortars [5, 6]. Mussel shells have been studied as a substitute for coarse and fine natural aggregates in concrete mixes. There is no trace of such research topics for the Italian mussel shell industry. In the following work, preliminary laboratory tests were carried out on both Orosei marble and mussel shells to determine the characteristics of these materials. Various cement mixtures were made, replacing Orosei marble and mussel shells in various percentages.

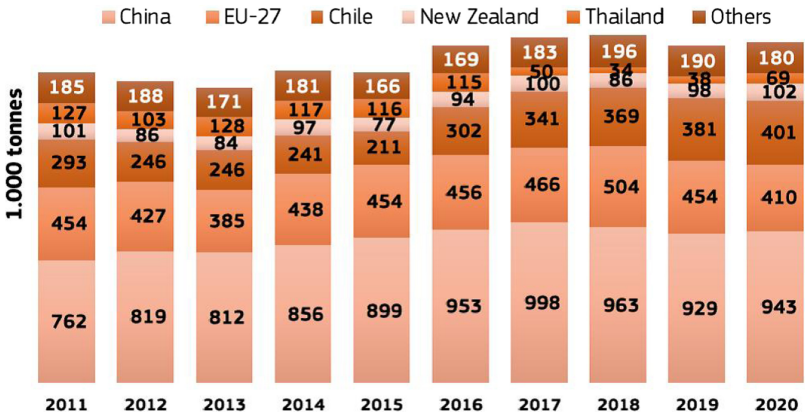


Fig. 1. Global mussel production 2011–2022 source FAO (Food and Agriculture Organization).

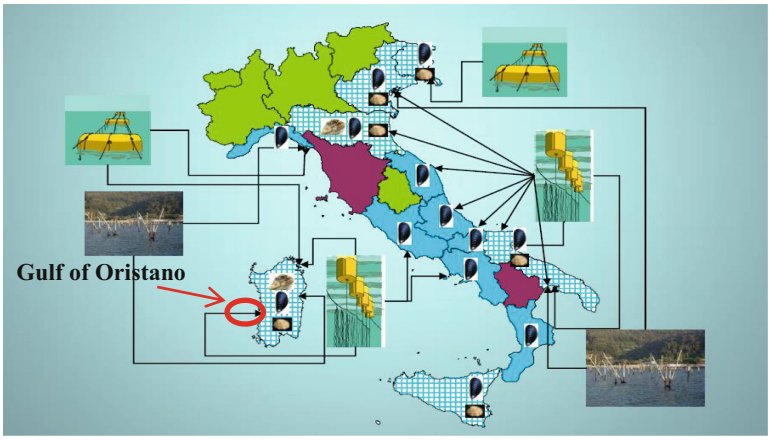


Fig. 2. Main mussel shells aquaculture production in Italy (Source ISPRA, Institute for Environmental Protection and Research).

Obtaining the correct mix design using high percentages of marble and mussel shells by-products by understanding the best substitution rate of each size fraction and maintaining good mechanical and physical performances is the main goal of the research.

The paper is structured as follows. Section 2 describes the materials and methods followed in this research. Section 3 presents the tests carried out in the laboratory on marble by-products and mussels shells. The mixes with the alternative materials prepared in the laboratory were reported in Sect. 4. Section 5 illustrates the potential applications, limits of this research, and future studies. Finally, Sect. 6 provides the conclusions of this research.

2 Materials and Methods

The Orosei marble used in this research comes from a carbonate formation that develops in correspondence with Tuttavista Mount (see Fig. 3), a region of Orosei, located on the east coast of Sardinia. The sedimentary rock that makes up the carbonate formation is classified as biopelsparite, i.e. a limestone with a prevalent structure of very fine-grained calcium carbonate (calcite, dolomite, micro and cryptocrystalline minerals) within which they are distributed, in different ways from area to area, particles such as fossils, shells and remains of organisms and fragments of other carbonate minerals. It is classified as a colorful non-crystalline marble, with a high percentage of calcium carbonate. The production of Orosei marble has produced around 3 million cubic meters of scraps in Sardinia with characteristics of excellent mineralogical and technological quality and different granulometry (0.001 mm – 2000 mm). The marble by-products were used to replace the coarse natural aggregate fraction. In this work, the following tests were performed on marble scraps: Particle Size Distribution (PSD) analysis, Atterberg Limits (AL), and Los Angeles Abrasion (LAA) test.



Fig. 3. View of Orosei marble district.

The other by-products used in this research are mussel shells that are considered waste materials of the maritime industry and a large part ends up in landfills, generating millions of tons of waste every year. They are mainly composed of calcium carbonate (90–95%) [5–7].

The shells are inorganic and of calcareous composition, incompatible with the accelerated composting process. They mainly end up in landfills or at the bottom of the sea,

thus producing an important environmental impact. This work investigated the performance of mussel shells as an aggregate in concrete as a partial replacement of natural fine aggregates.

To prepare the mixes, Portland Cement (PC) was used, type CEM II 42.5R, 12% by weight of the aggregates and water 9% by weight of aggregates. The mussel's by-products before any utilization were treated at 135 °C for 30 min [2], using the treatment required by European regulations for poultry, feeding as a reference [7]. This procedure ensures the disinfection of the aggregates and warrants their safe handling and storage. After the treatment, the mussel by-products were crushed and screened obtaining a fraction 0–4 mm. Finally, the mixes were studied for non-structural concrete (NSC) for a total of n°6 mix designs. For the first n.5 mixes six cubic samples (150 × 150 × 150 mm) were prepared and cured at 14 days, while for the mix n.6 specimens were cured and tested at 14 and 28 days before undergoing compressive strength.

3 Laboratory Test

The first material tested was the Orosei marble by-product. The aggregates were collected from stockpiles according to UNI EN 932–1 [8] (Italian organization for standardization and specifications and methods of testing). In the laboratory, they were reduced (see Fig. 4) according to UNI EN 932–2 [9] specifications to perform physical and mechanical tests. Preliminary tests carried out consisted of the determination of PSD according to UNI-EN 933–1 [10], AL according to CNR UNI 10014 [11], and the LAA test according to UNI EN 1097 2 [12]. To perform the tests the Orosei marble was dried in hoven at 105 °C as shown in Fig. 5 and, once cooled, they were quartered. A quarter of the material was used for the PSD and a quarter for the AL. For the AL, a quantity of material passing through 0.42 mm was used. The plastic limit is determined by measuring the minimum water content at which the material can be shaped into sticks with a 3 mm diameter until it starts to crack. It has been found that the material is not plastic. In Fig. 6 is



Fig. 4. Laboratory sample of marble by-product.

reported the PSD of the marble by-products that passes the 16 mm sieve. According to the classification UNI EN 10006 [13] the material resulted in class A1-a.



Fig. 5. Sample of marble by-products in the oven to be dried.

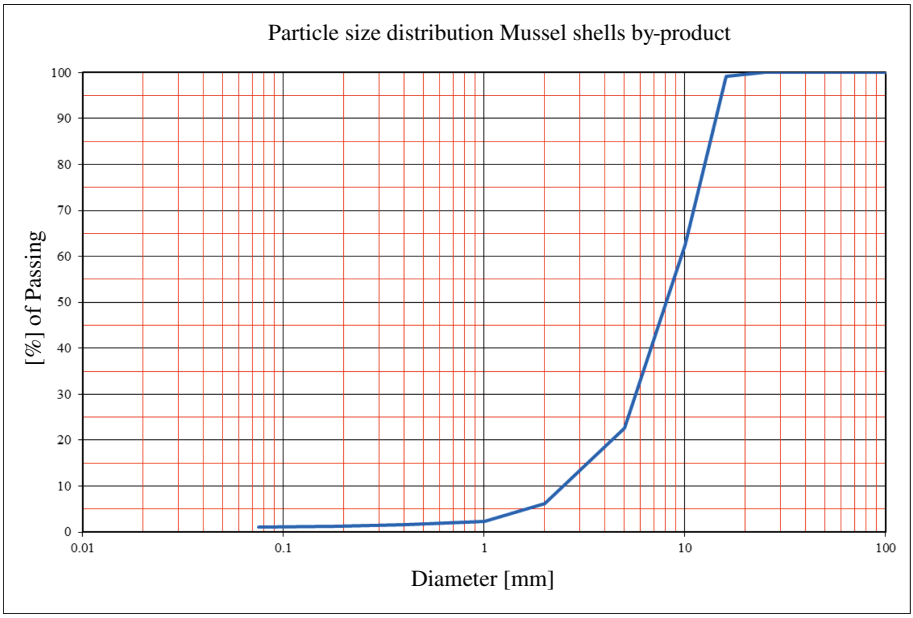


Fig. 6. Particle size distribution of marble by-products.

The other test conducted on Orosei marble was the LAA test. The LAA test measures the weight loss due to the fragmentation of a stone aggregate subjected to rolling action

with metal spheres in a special cylinder. The sample was prepared for the test according to the indications in the UNI EN 1097-2 technical standard (see Fig. 7). The sample was initially screened, washed, and dried in the oven. The test portion used weights of 5000 g. The sample and the spheres were introduced inside the cylinder. The machine was made to complete 500 revolutions at a constant speed between 31 and 33 rpm. At the end of the test, the material was washed and passed through a 1.6 mm sieve, and the portion retained on the 1.6 mm sieve was dried.



Fig. 7. Marble by-products being washed.

The LAA coefficient was calculated using the Eq. (1):

$$LA = 5000 - \mathrm{M}/50 \tag{1}$$

M: the mass retained on the 1.6 mm sieve, in grams.
The test results are shown in the following Table 1.

Table 1. Physical and mechanical properties of marble by-products aggregates.

Sample n.	Particle density (g/cm^3)	Los Angeles Abrasion (%)	Atteberg Limit
1	2.62	24.47	Non-plastic
2	2.63	24.50	Non-plastic
3	2.63	24.48	Non-plastic

From this preliminary data, the material is non-plastic and with a low value of LAA, all characteristics good to be used in the road industry.

Based on the chemical analysis that has been conducted it can be said that the composition of the Orosei marble has a 97% of calcium carbonate ($CaCO_3$) [14].

The other tested material was the mussel shells (see Fig. 8). The main component of mussel shells is calcium carbonate, approximately at a percentage of 95% [2].



Fig. 8. Mussel shells before any treatment.

The mussel shells before any testing procedure were placed in a hoven and treated at 135 °C for 30 min. This procedure is necessary to ensure the disinfection of the aggregates and warrants their safe handling and storage. Subsequently, the mussels shells were crushed and during crushing separated from the byssus (see Fig. 9). The byssus is the bundle of filaments secreted by mollusc that function to attach the mollusc to a solid surface. To perform the crushing a LAA machine was used with n. 11 steel spheres and a time of rotation of 30 min. This type of procedure ensured the possibility of obtaining a good fine fraction of the shells and also the possibility of removing the byssus attached to the mussels. On the obtain fine fraction of crushed mussels PSD tests were performed. In Fig. 10, is reported the chart of the PSD, in which more than 90% passes the 5 mm sieve. For the research only the fraction 0–4 mm was used for the mixes, for this reason, sieving was performed to eliminate the fraction above the 5 mm sieve. Also, an AL test was performed and as expected the aggregates resulted in non-plastic. According to the classification UNI EN 10006 [13] the material resulted in class A1-b and so, a good material for road construction purposes.

From this preliminary characterization of the marble and mussel shells by-products, the research focused the attention on the potential application of using such materials in plain concrete mixes. For this reason, n.6 mixes were prepared, cured, and tested to evaluate mechanical performances. More specifically, the marble by-products partially replaced the coarse natural aggregates. The crushed and sieved mussel shells totally or partially replaced the fine natural aggregates.



Fig. 9. Mussel shells crushed and separated from the byssus.

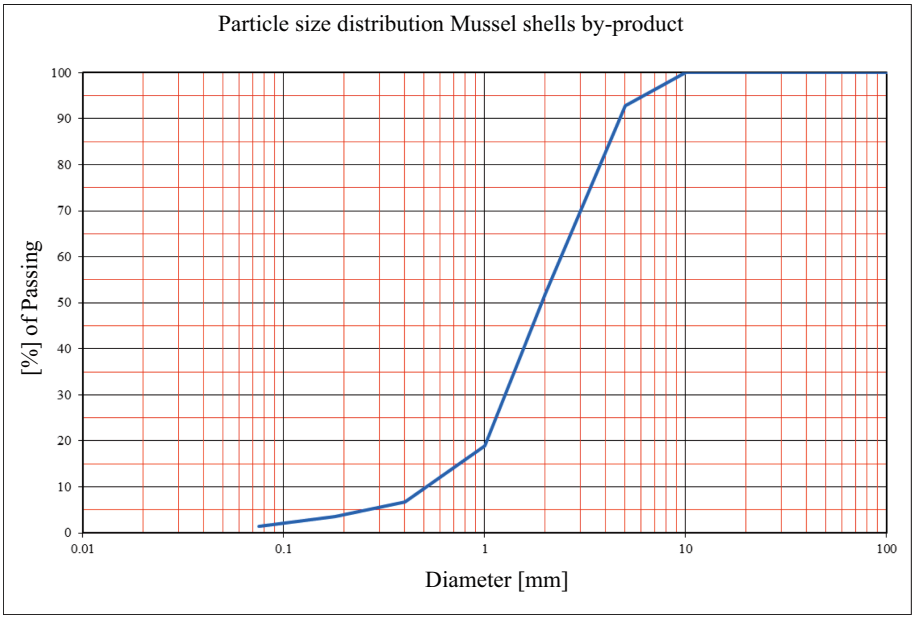


Fig. 10. Particle size distribution of mussel shells by-product.

4 Materials and Plain Concrete Mixes

In the first phase, a total of n.5 mixes were prepared, varying the substation rates of each natural aggregate size fraction (fine or coarse) using marble and mussel shells by-products. The cement used was a CEM II 42.5R, and no type of additive was introduced. A natural gravel coarse aggregate of 2–10 mm fraction was used. The water-cement (w/c) ratio used was 0.75, corresponding to 9% of water by weight of the aggregates and 12% cement by weight of the aggregates. The lowest w/c recorded in the literature was 0.3 [15] whereas the highest recorded w/c ratio was 0.8 [16] (see Fig. 11).



Fig. 11. Preparation of one of the mixes.

In this first series, no fine natural aggregates were used. Natural coarse gravel was partially replaced by Orosei marble by-products using 3 substitution rates 80%, 70%, and 50% (by weight), while mussel shells were used as fine aggregates. In Fig. 12 are shown different plain concrete mix samples, ready to be tested.

The following mixes were prepared as shown in Table 2.

The compressive strength test of the concrete specimens was carried out according to the UNI EN 12390–3:2003. The test was performed on each sample at 14 days of curing. The average results obtained are shown in the Table 3.

From the preliminary tests conducted, it can be seen that the concrete obtained from mix n.1 can be qualified as structural concrete. It must be said that structural concrete must have a value of compressive strength $\geq$ 20 MPa, while NSC must have a value of at least 10 MPa.

In this preliminary set of tests, segregation appeared during the granular aggregate mixture, especially in mix n.1 and mix n.2 attributed to the lack of fine aggregates in the mix design.

On the basis of these preliminary results, a n.6 mix was studied and tested. It consisted of cement type CEM II 42.5R, additive was used a super-plasticizer based on polycarboxylate, to give fluidity to the mix and also increase workability.



Fig. 12. Different mixes of plain concrete cubic samples ready for testing.

Table 2. Plain concrete mixes design.

Mixes n.	Orosei Marble (%)	Natural Gravel Coarse Aggregates (%)	Mussel Shells (%)
1	80	20	0
2	80	15	5
3	70	10	20
4	70	0	30
5	50	0	50

Table 3. Average Compressive strength results.

Mixes n.	Orosei Marble (%)	Natural Aggregates (%)	Mussel Shells (%)	Compressive Strenght (MPa)
1	80	20	0	20.92
2	80	15	5	15.93
3	70	10	20	11.77
4	70	0	30	10,67
5	50	0	50	7,47

The w/c ratio used was 0.50.

Regarding the aggregates used, natural gravel coarse aggregate fraction of 8–16 mm and 4–8 mm fraction was introduced in the mix, while a silica sand fraction of 0–4 mm was also used. In this mix, the marble by-products were used to partially replace 50% by weight of the fraction 8–16 mm coarse natural aggregate, while the crushed mussel shells were used to partially replace 19.5% of the fraction of silica sand fraction 0–4 mm. In Table 4 is reported the mix design composition for mix n.6.

Table 4. Plain concrete mixes.

Mix n.	Orosei Marble By-product 8–16 mm (%)	Natural Gravel Coarse Aggregates 8–16 mm (%)	Natural Gravel Coarse Aggregates 4–8 mm (%)	Natural Silica Sand Aggregates 0–4 mm (%)	Fine Mussel Shells by-product 0–4 mm (%)
6	14.3	14.3	29.55	33.81	8.1

For the mix n.6 compressive tests were performed at 14 and 28 days of curing. Two sets of specimens were cast and cured at 14 days consisting of n.3 specimens and n.3 specimens were cured at 28 days and tested. In Table 5 are reported the average compressive strength results at 14 and 28 days of curing.

Table 5. Average Compressive strength results.

Mixes n.	14 days curing Comprresive Strenght (MPa)	28 days curing Comprresive Strenght (MPa)
6	6.2	10.2

It can be seen from the results that mix design n.6 reaches the minimum value of compressive strength resistance to be considered NSC. It was seen from the test conducted and during the preparation of the mixes that the presence of fine mussel shells in concrete mixes: workability, density, and compressive strength of concrete decrease as the percentage of seashells increases, this is known in literature [2].

5 Potential Applications and Future Studies

The potential application of mussel shells by-products as substitute aggregate in concrete can be considered a valid alternative to improve various properties of concrete. The use of mussel shells by-products in concrete could provide environmental benefits in different ways: introduce a circular economy approach to the waste management of mussel shells, and reduce the exploitation of natural aggregate leading to a reduction of CO_2 emissions.

Also, the use of mussel seashells aggregates could provide an economic advantage in the construction industry since seashell by-products are generated in large quantities and have lower commercial value than natural aggregates. Possible implications of seashell aggregate concrete in construction are illustrated below.

Reduction in the density of concrete due to the incorporation of mussel seashells aggregates could be beneficial in special applications where lower self-weight of concrete is needed and not great strength is required. The possible application of seashell aggregate concrete in road pavements subjected to light traffic volume will depend on the mechanical properties (especially hardness) of the seashells, as well as the skid resistance and abrasion resistance of seashell aggregate concrete.

Current research shows that seashell aggregate has restricted potential in high-performance applications such as structural concrete and concrete exposed to aggressive environments. This is due to the presence of salts and organic impurities in seashells as well as reduced workability and lower strength of seashell aggregate concrete. In future applications, proper cleaning is needed by washing the shells and also to improve workability superplasticizers are essential. At present there are no studies on using mussel shells aggregate in reinforced concrete for this reason there is a need to study the structural behavior of reinforced concrete slabs, beams, and columns manufactured with treated seashells as partial replacement aggregate at different substitution levels and mix ratios is needed.

6 Conclusions

In this work, the potential use of Orosei marble scraps and mussel shells by-products was examined. The physical and mechanical properties of the material were analyzed and various concrete mixtures were prepared, with different percentages of substitution. Finally, the mechanical performances of each mixture were evaluated. Initially, five mixes were prepared in which coarse aggregates were replaced by marble by-products, and mussel shells totally replaced fine aggregates. In these mixtures with higher percentages of mussel shells, workability decreases, this behavior is known in the literature. In fact, the decrease in workability is owing to the irregular shape of the particle, which promotes particle friction, and the increased surface area, which increases the demand for water, as well as water absorption due to internal voids in seashell debris [17]. Also, it must be said that no super-plasticizer was used in the preliminary tests, which could have helped the fluidity and workability of the mixes. In all the compressive tests conducted in which mussels shells by-products were introduced in the mixes, all values were under 20 MPa. While mix n.2, n.3 and n.4 had compressive strength values that could be suitable for NSC.

In mix design n.6 marble by-products were used to partially replace 50% by weight of the fraction 8–16 mm coarse natural aggregate, while the crushed mussel shells were used to partially replace 19.5% of the fraction of silica sand fraction 0–4 mm. An increase in the fluidity of the mix was obtained by introducing a super-plasticizer. Also, no segregation appeared in this mixed design. The compressive strength value obtained at 28 days achieved the value of 10.2 MPa suitable to be considered a NSC.

From the preliminary tests conducted, it can be said that by-products of marble and mussel shells can be considered a valid alternative to partially replace natural aggregates. Further studies are needed to better understand such materials especially evaluate the more suitable percentage of mussel shells by-products in substitution of natural aggregates. Considering the data from the literature review and the data obtained in this research optimum percentage replacement of fine natural aggregates must not exceed 20%.

The reuse of these materials is essential in a sustainable and environmentally friendly circular economy approach. The resulting advantages are less exploitation of natural resources, less waste production, and less CO_2 consumption.

Acknowledgements. This study is supported by the projects "WEAKI TRANSIT: WEAK-demand areas Innovative TRANsport Shared services for Italian Towns (Project protocol: 20174ARRHT_004; CUP Code: F74I19001290001), financed with the PRIN 2017 (Research Projects of National Relevance) program and e.INS Ecosystem of Innovation for Next Generation Sardinia - SPOKE 8 - (CUP F53C22000430001 –MUR Code: ECS00000038) financed with PNRR (National Recovery and Resilience Plan). We authorize the MIUR to reproduce and distribute reprints for Governmental purposes, notwithstanding any copyright notations thereon. Any opinions, findings and conclusions or recommendations expressed in this material are those of the authors, and do not necessarily reflect the views of the MIUR.

Author Contributions. Concept and methodology, Rombi, J., Salis M. and Coni, M.; experimental campaign and validation, Rombi, J., Salis M. and Olianas M., analysis, Rombi, J., Salis M., and Maltinti F., writing, review and editing, Rombi, J., Salis M. and Maltinti F., project administration, Coni, M.. All authors have read and agreed to the published version of the manuscript.

References

1. Careddu, N., Siotto, G.: Promoting ecological sustainable planning for natural stone quarrying. The case of the Orosei marble producing area in eastern sardinia. Resour. Policy **36**(4), 304–314 (2011)
2. Martínez-García, C., González-Fonteboa, B., Martínez-Abella, F., Carro-López, D.: Performance of mussel shell as aggregate in plain concrete. Constr. Build. Mater. **139**, 570–583 (2017)
3. EUMOFA, European Market Observatory for Fisheries and Aquaculture Products, Mussels Shells in EU, European Commision, pp. 1–91 (2022)
4. ISPRA, Institute for Environmental Protection and Research. https://www.isprambiente.gov.it/files2018/eventi/cadeau/Giovanardi_ISPRA_MolluschicolturainItalia. Report (2018)
5. Martínez-García, C., González-Fonteboa, B., Carro-López, D., Martínez-Abella, F., Juan Luis Pérez, O.: Characterization of mussel shells as a bio-based building insulation material. In: The 3rd International Conference on Bio-Based Building Materials, Proceedings, pp. 526–531. Belfast (2019)
6. Uchechi, G.E., John, C.E., Bennett, I.E.: Properties of seashell aggregate concrete: a review. Constr. Build. Mater. **192**, 291–299 (2018)
7. Parliament, European. Regulation (EC) No 1069/2009 of the European parliament and of the council. 21 October 2009. Laying down healt rules as regards animal by-products ande derived products not intended for human consumption and repealing Regulation (EC) No 1774/2002 (Animal by products Regulation)

8. UNI EN 932–1. Tests for general properties of aggregates - Methods for sampling. Italian Organization for Standardization (U.N.I.)
9. UNI EN 932–2. Tests for general properties of aggregates - Methods for reducing laboratory samples. Italian Organization for Standardization (U.N.I.)
10. UNI EN 933–1. Tests for geometrical properties of aggregates - Determination of particle size distribution - Sieving method. Italian Organization for Standardization (U.N.I.)
11. CNR-UNI 10014. Determination of the Consistency (Atterberg) of a Soil. Italian National Research Council (C.N.R.) and Italian Organization for Standardization (U.N.I.)
12. UNI EN 1097 2. Tests for mechanical and physical properties of aggregates. Methods for the determination of resistance to fragmentation (2010)
13. UNI 10006. Road construction and maintenance - Technical provisions for use of soils (2002)
14. file:///C:/Users/james/Downloads/Materiali%20lapidei.pdf (2011)
15. Nguyen, D.H., Boutouil, M., Sebaibi, N., Leleyter, L., Baraud, F.: Valorization of seashell by-products in pervious concrete pavers. Constr. Build. Mater. **49**, 151–160 (2013)
16. Falade, F., Ikponmwosa, E.E., Ojediran, N.I.: Behavior of lightweight concrete containing periwinkle shells at elevated temperature. J. Eng. Sci. Technol. **5**(4), 379–390 (2010)
17. Mo, K.H., Alengaram, U.J., Jumaat, M.Z., Lee, S.C., Goh, W.I., Yuen, C.W.: Recycling of seashell waste in concrete: a review. Constr. Build. Mater. **162**, 751–764 (2018)