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On the use of seashells as green solution to mechanically stabilise dredged sediments

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ABSTRACT

The article reports the results of an experimental activity conducted on dredged fine-grained marine sediments and aimed to find out novel eco-friendly solutions for their mechanical stabilisation. The main idea of this research is to use seashells, i.e., another waste material, to partially replace cement binders in the mechanical stabilisation of sediments for the production of a new stable material that can potentially be used in construction. To this aim, an original procedure has been developed to obtain a powder of mussel shells without their calcination. Physical properties, one-dimensional compression behaviour and permeability of the novel mixtures including sediments, mussel shell powder and cements are presented for different curing times. The efficacy of the solutions is assessed also by comparison with the performance of control mixtures prepared by mixing the same sediments with cement only. The effects of the different treatments on the soil properties were analysed, demonstrating multiple beneficial effects of using the mussel shell powder. Evidence is provided that seashells represent a viable alternative to cement, as they were found to be as effective as traditional hydraulic binders, when replacing them up to ¼, in enhance geomechanical and geochemical performance of the stabilised material.

Keywords: dredged sediment; mussel shells; mechanical stabilisation.

1. Introduction

The sustainable use of resources is a fundamental aspect in a world where there is an exponential increase in the demand for them to sustain the constant economic and societal growth. This requires new strategies that have to be centred on the transformation of waste into recycled resources for use in other applications. This approach, inspired by the fundamentals of the circular economy, can be applied to different fields of Civil and Environmental Engineering, including the management and the reuse of marine sediments which are dredged periodically (in Europe about 100-200 million m³/year) in coastal areas for port development and/or for site remediation (SedNet, 2011). In this context, in the last 30 years, many research contributions in geotechnical engineering focused on the mechanical stabilisation of sediments by commercial binders such as cements and lime (e.g., Sherwood 1957; McDowell 1959; Taylor and Orman 1960; Mitchell and Hooper 1961; Dumbleton 1962; Croft 1967; Mitchell 1981; Federico et al., 2015). However, despite the concrete production could reach 5 billion tonnes by 2030 (WWF 2019), its manufacture process is still environmentally harmful for the related large CO₂ consumption and the release of pollutants: for every ton of clinker produced, one ton of CO₂ is released into the atmosphere. For this reason, in the most recent years, some solutions have been proposed making use of more sustainable binders that reduce the CO₂ consumption and accomplish with the so-called green

new deal (Yoobanpot et al., 2018; Roque et al., 2022; Paleologos et al., 2022; Petti et al., 2023). The present study intends to contribute to this field of research through the development of a novel sustainable solution for the stabilisation of sediments. The new binders have been obtained by partial substitution of the commercial ones with an originally prepared powder by recycling mussel shells. They are a particular food waste made up of 95% calcium carbonate, that makes shells non-biodegradable, difficult to compostable and of a fairly long disposal. The global production of marine bivalves for human consumption is more than 15 million tonnes per year (Wijsman et al. 2019), which is about 14% of the total marine production. Moreover, according to numbers of Fao-Fishstat (FAO 2016), Europe is the second worldwide producer of mussels. It follows that seashells represent a waste that is currently produced in large quantities in aquaculture and dumped in landfills with complex and expensive disposal procedures.

This is why recent research studies (Papadimitriou et al., 2017; Morris et al., 2019; Uster et al., 2014) focus on the use of mussel shells for numerous applications from the cosmetic to the fertiliser industry and as adsorbents of contaminants in soils. The study presents the results of laboratory tests carried out on both the novel mixtures including sediments, different types of cements and the mussel shell powder and control mixtures prepared by using the same sediments and cements only. Moreover, data on mixtures obtained by mixing sediments with the same cements and same quantities of powder of quarry limestone, as primary source of Calcium carbonate, are

shown for comparison. After having presented the results relevant to the base materials and the research programme, the results of the strength of the mixtures are explored by using the texturometer device and computing the consistency index on the different mixtures after 28 days of curing.

2. Materials and methods

2.1. Untreated sediment

The untreated sediment (US, Fig. 1 a-b) has been dredged from the Taranto port in the south of Italy. As summarised in Table 1, US is clayey silt with low sand fraction. As suggested by both standards and literature (e.g., BS 1337; Sollecito et al., 2019), the natural water content, w_0 , and the Atterberg limits were corrected to take account of the pore fluid salinity. The sediment is characterised by low value of the unit weight of soil solids ($\gamma_s = 15.95 \text{ kN/m}^3$) and TOC = 1.52 (Table 1) value are derived as total carbon content minus TIC. Such organic material is essentially represented by algae and is retained to the ASTM sieve n. 40 (Fig. 1 c). This evidence has brought about a deeper investigation of the Atterberg limits that were determined on both the total sample, following an unconventional procedure (UP; full square in Fig. 2), and on the material passing the ASTM sieve n. 40, according to the ASTM procedure (empty square in Fig. 2). The Figure shows that when the limits are determined on the total sample both liquid limit and plasticity index increase (i.e., $w_{LUP} = 53\%$; $PI_{UP} = 28\%$; $w_{LASTM} = 46\%$; $PI_{ASTM} = 23\%$), so that the US passes from being classified as CL to CH soil (USCS, ASTM D2487-17e1).

Table 1. Physical properties.

w_0 [%]	SF [%]	MF [%]	CF [%]	γ_s [kN/m^3]	TOC [%]
ASTM D2216-4542	ASTM D422	ASTM D422	ASTM D422	ASTM D854-14	DIN EN 15936
74	5.8	63.8	30.4	15.95	1.52

Key. w_0 : water content; SF: sand fraction; MF: silt fraction; CF: clay fraction; γ_s : unit weight of soil solids per unit volume; TOC: total organic carbon

The distance between the two points in the Casagrande's plasticity chart is a first measure of the active role of these organic fractions in interacting with the clay particles to modify the sediment plasticity. The consistency index of the sediment ($CI = (w_L - w_0)/PI$) is always negative and equal to -0.73 and -1.2 if computed by using either the Atterberg determinations on the total materials or following the ASTM procedure, respectively.

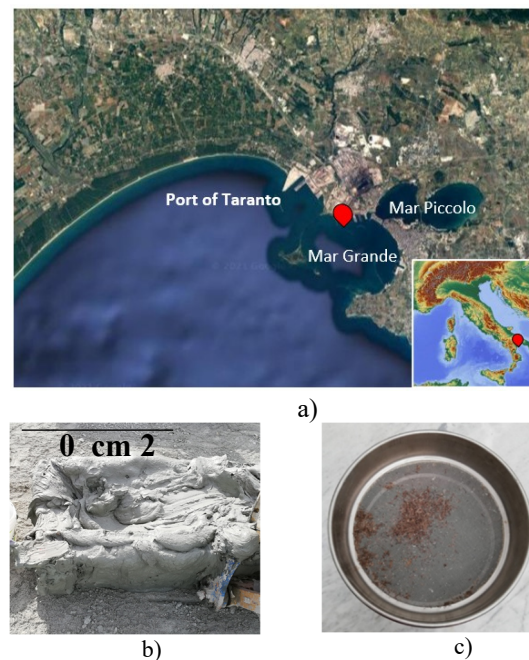


Figure 1. a) Port of Taranto and its location in the Mediterranean Sea. The red dot indicates the site of sampling of the studied materials; c) dredged sediment from the Port of Taranto; c) Organic component retained on sieve n. 40.

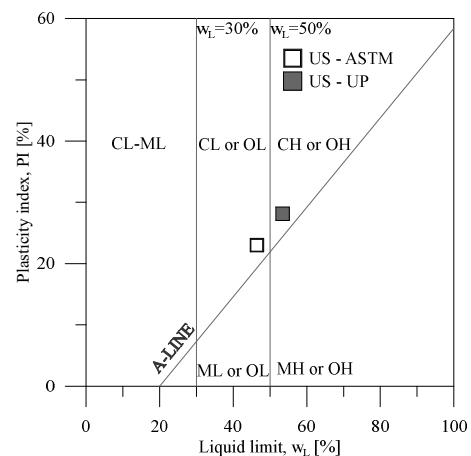


Figure 2. Casagrande plasticity chart of marine sediment from the Port of Taranto. US-ASTM: standard procedure, US-UP: unconventional procedure.

Thermogravimetry tests were also performed to explore further the nature of the sediment skeleton and its organic matter content, based on the main thermal reactions occurring within different temperature ranges (Dell'Abate et al., 2000; Lopez-Capel et al., 2005; Maharaj et al., 2007; Emmerich, 2011; Kristl et al., 2016; Sollecito et al., 2021). In the present study, the TG was performed using a METTLER TA SDTA851E system, on air-dried specimens, heated upon a temperature range of 25-1000°C. The resulting curves of the total mass loss (TG) and the derivative mass loss (DTG) are shown in Fig. 3, respectively, for the dredged sediment sample under study.

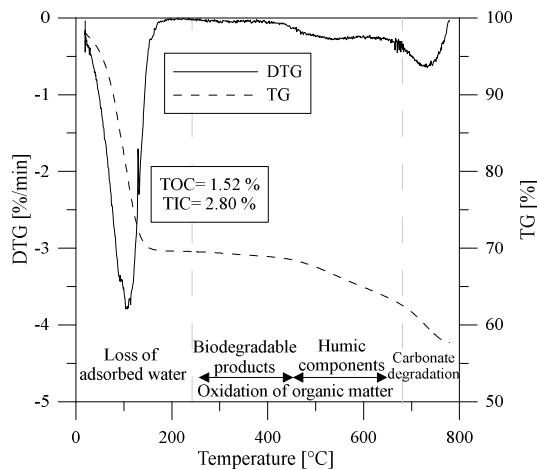
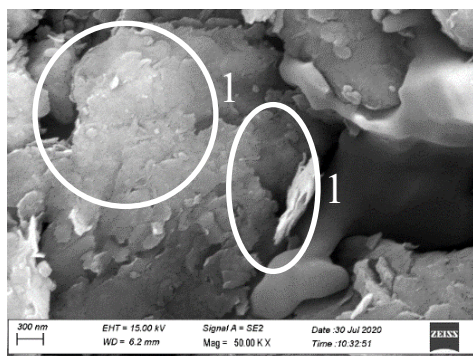


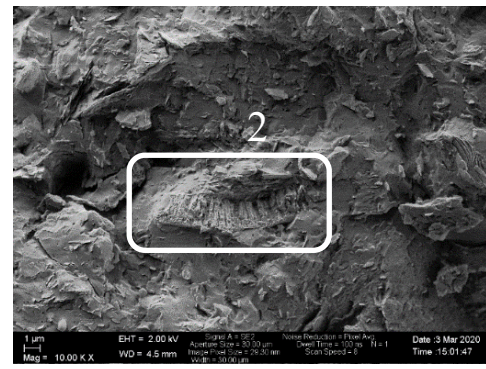
Figure 3. TGA and DTG curves of marine sediment.

At temperature values about 200°C, the soil has experienced the loss of adsorbed water and the dehydration of both the clay aggregates and swellable clay minerals. Consistently, the DTG curves of the sample show a peak in the rate of mass loss at about 100°C (Fig. 3). In the temperature range 220–620°C, although the soil loses mass due to the dihydroxylation of the clay minerals (e.g., for kaolinite at 560°C, for smectite at 500–700°C), a part of the mass loss may also be due to organic matter degradation. The weight loss in the lower part of this temperature range (200–450°C) is associated with the combustion of carbohydrates and poorly altered organic matter, while that in the range 375–500°C is consequent to the degradation of more complex organic substances, such as humus, lignin, and aromatic compounds. Given this explanation, the DTG peak recorded in the sample at about 500°C also reveals the presence of humic components. For heating temperatures higher than 620°C, the mass loss of the soil is related to the process of carbonate thermal decomposition. At these temperatures, a peak is recorded in the DTG curve of the sediment sample.

The mineralogical composition, determined by XRD analyses, within the clay fraction of the US is essentially represented by smectite and mica which are predominant over chlorite and kaolinite. NaCl (halite) is also detected as result of the salt precipitation after the drying of the samples (Petti et al., in prep.). Finally, SEM photographs of the sediments reveal the typical illite clay plate morphologies in their face-to-face contacts (1 in Fig. 4) and highlight the presence of fossil prints (2) confirming the complexity of marine sediments (Vitone et al., 2020).



a)



b)

Figure 4. SEM photograph of the dredged sediment: 1 – contacts of clay particles; 2 – fossil prints.

2.2. Cements

Three types of cements were used: from the traditional type I Portland Cement 52.5R (P), to a more sustainable commercial binder, such as a type III blast furnace cement (T), to the most recent Sulphoaluminate cement (CS). Both T and CS are cement binders resulting from a Core process with less than 550 kg /t of CO₂ emissions and at least 30% of pre- or post-consumption recycled material. From the comparison of the data in Table 2, the main differences among the chemical composition of the three cements are the high percentages of SO₃ and Al₂O₃ in CS, high CaO content in P and CaO and SiO₂ in T.

Table 2. Chemical composition of the mussel shell powder, MS, and cement (Portland cement, P; blast furnace cement, T, Sulphoaluminate cement, CS).

Oxides [% mass]	T	CS	P	MS
CaO	44.99	40.19	63.06	53.60
Al ₂ O ₃	8.25	23.64	3.92	0.02
Fe ₂ O ₃	1.48	1.37	4.51	0
SiO ₂	32.81	7.57	18.09	0.038
MgO	5.79	2.75	1.59	0.239
Na ₂ O	0.52	0.89	0.5	0.355
K ₂ O	0.81	0.49	0.93	0.034
SO ₃	3.61	20.03	4.13	0.197
P ₂ O ₅	0.05	0.09	0.11	0.034
SrO	0.09	0.16	0.03	-
MnO	0.15	0.11	0.07	-
TiO ₂	0.38	0.35	0.22	0
Mn ₂ O ₃	-	-	-	0.002
Sr[PPM]	-	-	-	812
Ni[PPM]	-	-	-	48
Cu[PPM]	-	-	-	118
LOI	1.27	0.75	2.64	45.58

2.3. Limestone and mussel shells

In this study, the mussel shell powder, produced without calcination, is used in partially replacement of cement in mechanical treatment of sediment. To check its efficacy, as natural additive of cement, the performance of the mixtures of sediments, cements and mussel shell powder has been compared to that of mixtures prepared by adding to the same sediments and commercial cements, the same quantity of geogenic quarry-limestone powder (L). The limestone was crushed and sieved to

have two grain particle size distributions: i) L, that is characterised by $D_{50} = 2.84 \mu\text{m}$ and uniformity coefficient $C_u = D_{60}/D_{10} = 2.409$; ii) CL, coarser than the previous one, with $D_{50} = 9.38 \mu\text{m}$ and $C_u = 5.548$. D_{10} , D_{50} and D_{60} are the grain sizes corresponding to the 10, 50 and 60% of passing sieve, respectively.

The shells used in this study are of the type *Mytilus galloprovincialis* (Fig. 5). This type of seashell is an abundant waste in Europe and contains carbonates represented by calcite and a not negligible content of aragonite (Chilakala et al., 2019). To produce the mussel shell powder (MS), the following treatment was carried out: i) washing the shells with hot water for about 10 minutes and oven-drying at $105 \text{ }^\circ\text{C}$ for 48 hours to remove impurities from the shells (Othman et al. 2013); ii) milling the material in a Retsch jaw crusher; iii) sieving it to obtain two different particle size distributions, CMS and MS. The first one is characterised by $D_{50} = 127 \mu\text{m}$ and C_u equal to 0.754, the second one, MS, has instead $D_{50} = 6.32 \mu\text{m}$ and $C_u = 2.213$. Table 2 also provides evidence that the MS powder produced without calcination is essentially biogenic calcium carbonate (53.61 wt.% expressed as CaO) with small fractions of other oxides.

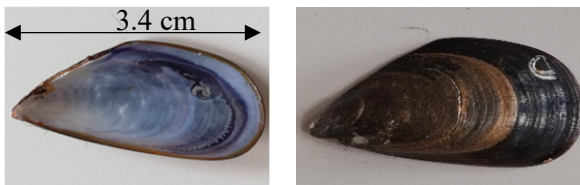


Figure 5. *Mytilus galloprovincialis* type of shell used in this study.

Fig. 6 shows the SEM analyses performed on the same type of mussel shells by Ballester et al. (2007). Differently from limestone of geogenic origin, where CaCO_3 aggregates consist mainly of rounded particles, the fabric of mussel shells is made up of elements of similar average size (2–6 μm) but more elongated shape. Moreover, as reported in the literature, the structure of mussel shells, like all bivalves, can be divided into three parts: the outer layer, periostracum, the middle layer, i.e., the prismatic layer (Gao et al., 2015), and the inner layer referred to as nacre (Martínez-García et al., 2017). Elongated shapes characterise both prismatic layer and nacre. This morphology, if compared to that of quarry-limestone, is evidently characterised by higher specific surfaces and then, it is likely more prone to chemical exchanges with the active components of both fine-grained sediments and cements.

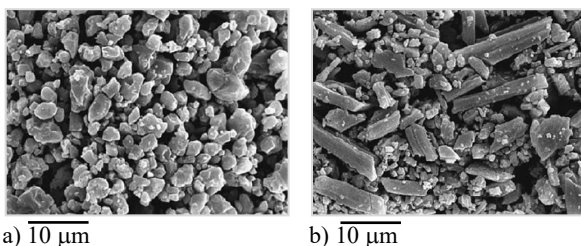


Figure 6. SEM images of (a) quarry limestone aggregate and (b) mussel shells (Ballester et al., 2007).

2.4. Experimental programme

The first part of the experimental programme entailed the comparison between mixtures formed by sediments, cements and limestone powder and the same mixtures where limestone was replaced by the mussel shell powder (MS). Both the powders were tested with the fine (i.e., MS and L) and coarser (CMS and CL) grain sizes. For each cement used (i.e., P, T or CS type), the sediment-mixtures were prepared by replacing 1/4 or 1/2 of cement with either mussel shell or limestone powder, for both finer and coarser-grained sizes. Finally, also traditional sediment-cement control mixtures were prepared and tested. The content of binders added to the sediment was adjusted according to the dry weight of the sediment, which was determined from its natural water content ($w_0 = 74.08\%$). The virgin material was first homogenised in a mechanical mixer (typical mixer for the mortars) for 10 minutes at medium speed (285 RPM). Subsequently, the binder was added, and the material was further mixed for 5 minutes at lower speed (140 RPM). As detailed in Table 3, after 7 and 28 days of curing in marine water, water content, Texturometer pressure probe and pH of curing water determinations were carried out on the mixtures. Sea water for the curing phase is chosen to use the same water that is naturally contained in the sediment. After curing phase, the sea water was removed, and the treated materials, without compaction, are tested. A first determination of the strength of the materials has been carried out by Texturometer apparatus (Fig. 7), i.e., a device commonly used in cement and construction industry and food industry for quick determinations about mechanical textural parameters (hardness, cohesiveness, viscosity, elasticity, adhesiveness, brittleness, chewiness, and gumminess). Although frequently used for cements, it has been here used on natural and treated sediment samples for a first assessment of the material strength. The test is based on the penetration of a cylindrical standard probe at controlled speed (i.e., 20 mm/min) for a defined length (40 mm). The probe in this case is a cylindrical probe with a diameter of 25.4 mm (or needle probe with a diameter of 1.8 mm for mixtures with more consistency, i.e., all mixtures with P and T cements).

3. Results and discussions

3.1. Comparison of L and MS sediment-mixtures

The results of Texturometer tests in term of peak pressure probe in 40 mm of penetration (Fig. 8a and 8a') and CI values (Fig. 8b and 8b') of sediments treated with L or MS and with P (Fig. 8a and 8b) or CS-cements (Fig. 8a' and 8b') are compared as determined after a 7 days of curing. As an example, the mixture indicated as US-6P-2CMS in Fig. 8a was prepared by adding to the untreated sediment (US) 6 wt.% of Portland cement (6P) and 2 wt.% of coarser mussel shell powder (2CMS), whereas US-6CS-2MS mixture (Fig. 8a') contains the dredged sediment (US) 6 wt.% of Sulphoaluminate cement (6CS) and 2 wt.% of finer mussel shell powder (2MS). Moreover, Fig. 8 shows the results obtained for

the relevant control mixtures, i.e., when mixing the same sediments with cement only in the same total percentage.

Table 3. Experimental programme for the mix design comparing the use of either MS or L powder in the SED-CEM mixtures. 7 and 28 days of curing time. Key. Text.: Texturometer test; pH: pH of curing water; w_0 : water content; US: untreated sediment; P: Portland cement; CS: Calcium sulfoaluminate cement; MS: mussel shell powder; CMS: coarse mussel shell powder; L: limestone powder; CL: coarse limestone powder.

Material	Test	
	Curing time	
	7 days	28 days
US-6P-2MS	Text., pH, w_0	Text., pH, w_0
US-6P-2CMS	Text.	-
US-6P-2L	Text., pH, w_0	-
US-6P-2CL	Text.	-
US-4P-4MS	Text., pH, w_0	Text., pH, w_0
US-4P-4CMS	Text., pH	-
US-4P-4L	Text., pH, w_0	-
US-4P-4CL	Text., pH	-
US-8P	Text., pH, w_0	Text., pH, w_0
US-6CS-2MS	Text., pH, w_0	Text., pH, w_0
US-6CS-2CMS	Text., pH	-
US-6CS-2L	Text., pH, w_0	-
US-6CS-2CL	Text., pH	-
US-4CS-4MS	Text., pH, w_0	-
US-4CS-4CMS	Text., pH	-
US-4CS-4L	Text., pH	-
US-4CS-4CL	Text., pH, w_0	-
US-8CS	Text., pH, w_0	Text., pH, w_0
US-6T-2L	-	Text., pH, w_0
US-6T-4L	-	Text., pH, w_0
US-8T	-	Text., pH, w_0

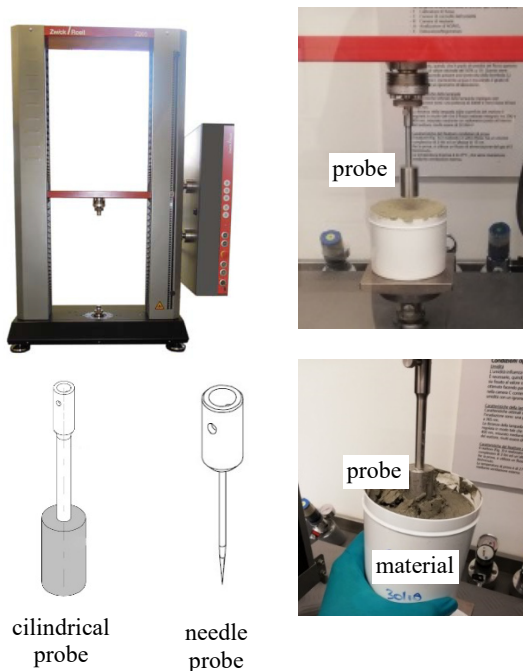


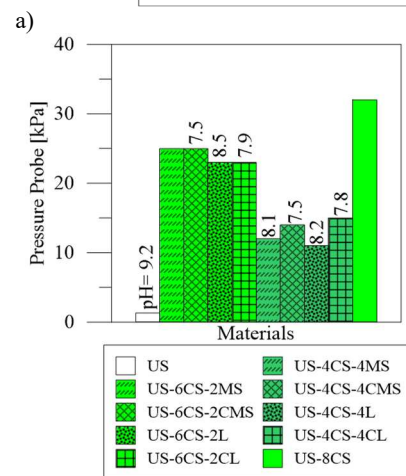
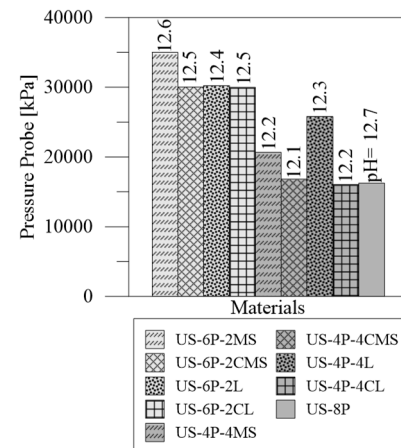
Figure 7. Photographs of the Texturometer device with the detail of the probe.

The pH values measured in curing water are also reported in the Figures.

The first general consideration that can be made is that P sediment-mixtures with 1/4 replacement ratio by MS or L show higher CI, pH, and pressure probe strengths than the corresponding ones with ratios 1/2 (i.e., 4% cement and 4% additive). All the mixtures including L or MS exhibit better performances than US-8P mixture. This is likely resulting from the effect that the mussel shells (and limestone) have on the hydration of cement compounds. In fact, the fine mussel shell powder (and limestone), in addition to filler effect, act as nucleation sites (the finer the powder, the better) promoting the precipitation of hydration products (Cyr et al., 2006). This accelerates the hydration process and would explain the higher strength measured at 7 days in the samples with treated with a binder in which part of the cement (2% of the overall 8%) was replaced with limestone or MS (Fig. 8).

Specifically, for P replacement ratios equal to 1/4 (i.e., 6% P and 2% additive), both MS and L-mixtures exhibit far higher values of CI and texturometer pressure than the relevant control mixture (US-8P).

Comparing the results relevant to the different types of cement used, it can be observed that CS-mixtures exhibit texturometer pressures, CI and pH values of curing water much smaller than the corresponding P-mixtures.



a')

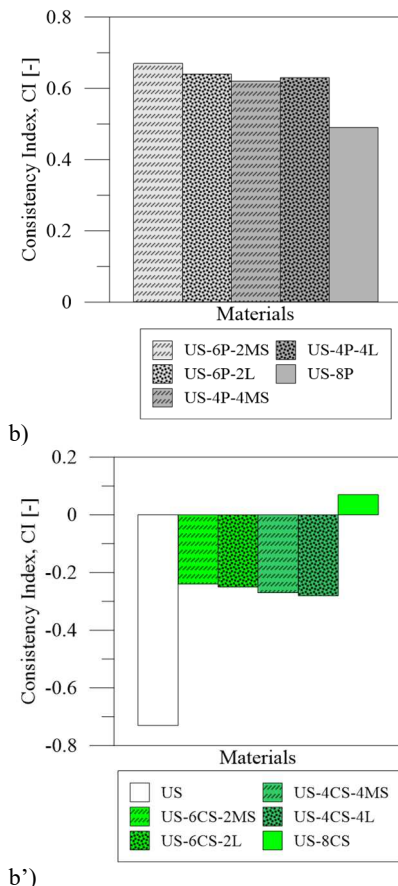


Figure 8. Pressure probe and pH values (number in the Figure) of P (a) and CS (a')-cement mixtures, CI of natural sediments (US) and P (b) and CS (b')-cement mixtures after 7 days of curing.

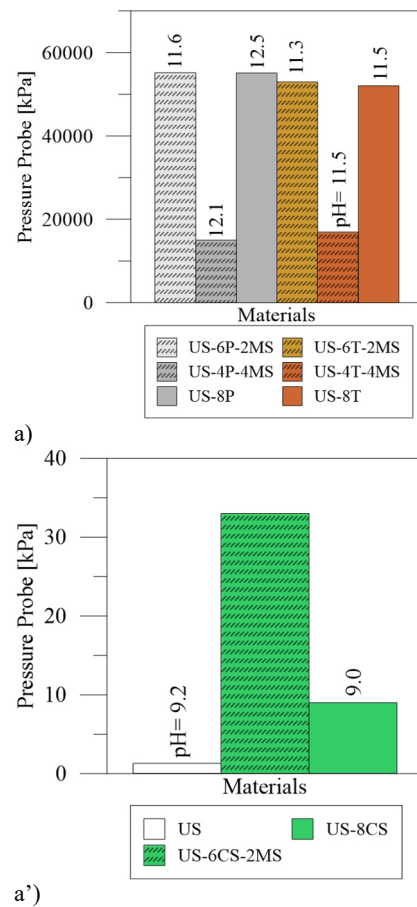
Specifically, higher average values of pH (about 12, ASTM D4972-01) have been measured for P-treated sediments, whereas pH values about 8 are typical of CS-sediment mixtures. Consistently, the CI values of the CS-mixtures remain negative except for the control mixture which has CI about zero (Fig. 8b').

Moreover, the partial replacements of CS with either L or MS seems to further reduce both texturometer pressure and consistency of the mixtures. However, the CS-mixtures exhibit texturometer strength and CI values always higher than those of the untreated sediment, US. It is relevant to note that, both consistency and texturometer strength of the sediment-cement mixtures including CMS or MS are generally either comparable or even higher than the corresponding ones with L. The better performance of mussel shell powder if compared to that of limestone, despite their similar composition, is likely due to the already-mentioned beneficial potential of laminated morphology that is peculiar of the shells. Moreover, treated sediments including MS or L finer powders with replacement ratio 1/4, exhibit higher or at least comparable strength than those with coarser additives, i.e., CMS or CL, irrespective of the type of cement. For 1/2 replacement ratios the opposite is recorded only when CS is used.

3.2. Consistency and texturometer strength of MS-sediment mixtures

For the sediment-mixtures exhibiting higher consistency and strength values after 7 days of curing, the effect of curing time was monitored by performing the same tests after 28 days of curing (Fig. 9). New determinations were also carried out on blast furnace slag (i.e., T) cement-sediment mixtures, i.e., US-6T-2MS and US-4T-4MS, and the corresponding control one (US-8T). Fig. 9 shows that the CI, texturometer strength and pH values of the T-cement mixtures are similar to those of the corresponding one obtained by using Portland cement. Moreover, longer curing times make the consistency and the texturometer strength increase for both the MS-mixtures and the control ones. As expected, CS-mixtures exhibit much lower performances. As far as the MS content is further increased to 1/2 replacement ratio, a decay in all the targeted parameters is recorded in the mixtures (Fig. 9).

Replacing 1/4 of the cement with MS not only does not seem to affect the results but it is found to cause an additional (albeit small) increase in consistency in the mixtures with P and T (3 and 5% for US-6P-2MS and US-6T-2MS, respectively). In contrast, the addition of MS does not appear to significantly change the performance of the sediment-CS mixture.



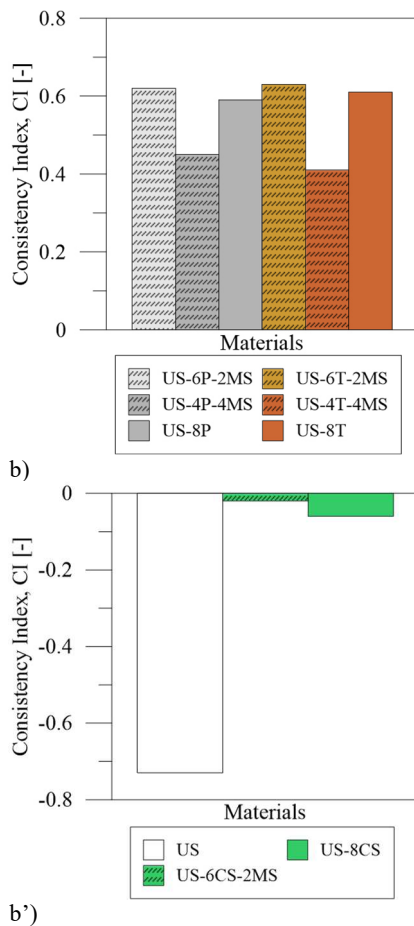


Figure 9. Pressure probe and pH values (number in the Figure) of P and T (a) and CS (a')-cement mixtures, CI of natural sediments (US) and P and T (b) and CS (b')-cement mixtures after 28 days of curing.

The results reported in Fig. 9a-a' show that the maximum pH values were exhibited by sediment mixtures treated with 8P and 8T (pH is about 12). The mixtures including CS show lower pH values, i.e., about 8-9).

All the stabilisation treatments studied induce a relevant increase in the consistency and/or pressure probe of Texturometer apparatus, despite the not negligible organic compounds present in the natural sediment (Fig. 3, Table 1). In this respect, Tremblay et al. (2002) show that, not only the quantity, but also the nature of the organic matter is relevant in affecting the effectiveness of soil mechanical stabilization with binders. For example, it is possible to mechanically stabilize by cement-binders an organic soil without a significant reduction in the final strength, if in the soil the organic compound is represented by humic components. On the contrary, strong acid compounds prevent the pore water from reaching a sufficiently high enough pH to develop cementing products for soils treated with 10% cement. In addition, oils and hydrocarbons are harmful to cement hydration because they coat the cement particles and delay the setting time. In the case of the sediments under study, the organic content essentially is represented by lignin and coal (humic components, Fig. 3), so that the results are in good agreement with those from the literature showing not significant inhibitory effects on mechanical stabilisation of soils (Tremblay et al., 2002).

4. Conclusions

The main idea of this research is to combine two waste materials (marine sediments and mussel shells) with cement binders to produce a new stable material that can potentially be used in many engineering applications.

In the paper, original experimental results on the mechanical performance of mixtures formed by sediments stabilised with three types of cement (i.e., commercial Portland and the blast furnace slag (T) and sulphoaluminate (CS) as two "greener" types of cement) and mussel shell powder are reported. Data are also shown for comparison of the same sediments when treated with control mixtures made up of cement only and cured up from 7 to 28 days.

The study provides evidence that the mussel shell powder, although non-calcinated, is a valuable partial substitute of cement in fine marine sediment stabilisation solutions. It can represent an interesting alternative that can reduce the consumption of natural resources, i.e., as crushed limestone, sand and gravel, extracted through highly impactful quarrying or river exploitation activities.

In fact, geomechanical data of mixtures produced by mixing sediment with binder (P or T) and MS show that replacement with mussel shells does not alter the performance of the mixture, which, despite the 25% reduction in binder content, exhibits physical-mechanical characteristics that are comparable or even better than those of sediment-binder mixtures. Specifically, the performance of the mixture MS-sediment-cement after 28 days of curing is comparable to that of sediment-cement both in terms of texturometer pressure and consistency index. When the Sulphoaluminate cement is used, low mechanical performance is recorded by all the mixtures. This could be caused by the nature of the hydration products of this type of cement and the presence of high amounts of soluble sulphates.

The performance of the original mixtures including mussel shells has been compared also to that of mixtures formed by the same cements, and powdered limestone, as primary source of calcium carbonate. The reported data appear to be consistent with each other and show that MS powder is sometimes even better performing than the same quantity of quarry-limestone powder. A possible reason for such result is presented as depending on the peculiar laminar morphology of the seashell microstructure.

Although these results have been obtained on a specific natural material, the marine sediment from an Italian site, and a specific type of mussel shell, it is the authors' contention that, for composition (silt with clay), medium plasticity, fluid consistency and high organic matter content of the sediment used and for the widespread presence of the *Mytilus Galloprovincialis* cultivations, most of the conclusions are applicable to other sediments as well.

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