MUSSEL SHELLS: A CANNING INDUSTRY BY-PRODUCT CONVERTED INTO A BIO-BASED INSULATION MATERIAL

Martínez-García, Carolina[1]; González-Fonteboa, Belén[2]; Carro-López, Diego[3]; Pérez-Ordóñez, Juan Luis[4]

[1]**PhD Student at the School of Civil Engineering**. Department of Civil Engineering, University of A Coruña. **Postal Address**: E.T.S.I. Caminos, Canales, Puertos. Campus Elviña s/n, 15071 La Coruña, Spain. **E-mail**: carolina.martinezg@udc.es. **Telephone number**: (+34) 881015463. **Fax**: (+34) 981167170

[2]**Associate Professor at the School of Civil Engineering**. Department of Civil Engineering, University of A Coruña. **Postal Address**: E.T.S.I. Caminos, Canales, Puertos. Campus Elviña s/n, 15071 La Coruña, Spain. **E-mail**: bfonteboa@udc.es. **Telephone number**: (+34) 881011442. **Fax**: (+34) 981167170

[3]**Associate Professor at the School of Civil Engineering**. Department of Civil Engineering, University of A Coruña. **Postal Address**: E.T.S.I. Caminos, Canales, Puertos. Campus Elviña s/n, 15071 La Coruña, Spain. **E-mail**: dcarro@udc.es **Telephone number**: (+34) 881015429. **Fax**: (+34) 981167170

[4]**Associate Professor at the School of Building Engineering**. Department of Civil Engineering, University of A Coruña. **Postal Address**: E.T.S.I. Caminos, Canales, Puertos. Campus Elviña s/n, 15071 La Coruña, Spain. **E-mail**: jlperez@udc.es. **Telephone number**: (+34) 881015434. **Fax**: (+34) 981167170

Abstract

The canning industry in Galicia produces 25 thousand tonnes of mussel by-product (i.e. unconsumed shells) per year, having a significant environmental impact. Mussel shell becomes suitable for different uses after undergoing heat treatment such as poultry feed or bedding, etc. However, other end uses for this by-product must still be explored. Building insulation material (for instance expanded polystyrene, extruded polystyrene, polyurethane, etc.) has a significant harmful effect on the environment, which is especially seen when analysing its entire life cycle (use of large amounts of energy and water for production, difficulty recycling or reusing waste in the event of demolition or refurbishment, etc.). The aim of this research is to prove the feasibility of using mussel shell as a building solution. Mussel shell has been found to have thermal and acoustic characteristics suitable for using it as building insulation material. Mussel shell confined inside an enclosed space (e.g. a wooden box) has a thermal conductivity similar to that of a light conifer wood, so it can be considered a material with low thermal conductivity. Furthermore, several acoustic studies indicate that a section of confined mussel shell displays a behaviour similar to that of commercialised insulation material. The embodied energy of mussel shell as a loose-fill material is similar to that of other bio-based insulation materials. The main goal of this paper is to demonstrate that it is possible to design mussel shells building solutions, that actively participate in environmentally responsive architecture.

Keywords:

Mussel shell; building insulation; thermal conductivity; sustainability; building acoustics

Highlights:

- Mussel shells aggregate can be used as loose-fill insulation material in buildings
- Placement of mussel shell as loose-fill material was tested in prototypes
- Mussel shell gravel has low thermal conductivity close to 0.10 W/(m.K)
- Mussel shell gravel has shown good acoustic behaviour

1 Introduction and objectives

The canning industry in Galicia produces 25 thousand tonnes of mussel by-product (i.e. unconsumed shells) per year (Caballero Míguez et al., 2009; Dias et al., 2011; Heinonen, 2014), which mainly ends up in landfill or at the bottom of the sea, having a significant environmental impact. This by-product is only 35 % (Iribarren et al., 2010; Xunta de Galicia, 2005) of the total cultured mussel production and is heat treated to become suitable for different uses (poultry feed or bedding, soil acidity correction and fertilizer). However, these uses are a minority, and other avenues need to be explored in order for 100 % of this by-product to be consumed.

Furthermore, the building sector is responsible for 40 % of the total energy consumed in Europe, as well as about 1/3 of greenhouse gas emissions for the entire planet (Asdrubali et al., 2015). For this reason, reducing energy consumption is one of the most important challenges for buildings in the future. Thermal insulation is undoubtedly one of the best ways to reduce energy consumption. The external envelope of a building plays a significant role as it strongly affects its surrounding microclimate (Galli et al., 2013; Pisello, 2015). In recent decades, development of this envelope has led to optimal thermal performance of vertical walls in terms of thermal transmittance. Furthermore, regarding the entire energy loss for buildings, a large amount of thermal energy is lost through opaque walls, so the use of adequately insulated walls has become essential (Asdrubali et al., 2016).

Nowadays, building material is expected to perform several functions, as well as be sustainable. For example, building material is expected to meet structural, thermal and acoustical demands (Glé et al., 2011).

Usually, thermal conductivity is used to define the thermal performance of a material. Thermal conductivity λ defines the steady state heat flow passing through a unit area of a homogeneous material, 1 m thick, induced by a 1K difference of temperature on its faces. It is expressed in terms of W/(m.K). There are different types of commonly used commercial thermal insulation materials: blanket (batts and rolls), foam board or rigid foam, loose-fill and blown-in, reflective system, rigid fibrous or fibre insulation, sprayed foam and foamed-in place, among others. It is well known that thermal conductivity depends on atomic and molecular structure, porosity, anisotropy, structural faults and defects of the material (Al-Ajlan, 2006). However, a material can be considered thermal insulation if its conductivity is lower than 0.1 W/(m.K) (Mati-Baouche et al., 2014), and most of the aforementioned commercial insulating materials are in the range of 0.04-0.05 W/(m.K).

Furthermore, acoustic comfort has become increasingly important for urban populations living in multi-occupant dwellings, as well as single family residential areas surrounded by busy roads, highways, high-speed railways, airports, etc. From an acoustic point of view, insulating material can be characterised in terms of its ability to counteract sound transmission and absorb impinging sound waves. Sound is dissipated inside the material by friction or thermal loss (porous materials), as well as due to the resonance phenomena (perforated and membrane absorbers) (Schiavoni et al., 2016). Regarding airborne sound insulation, this characteristic strongly depends on the mass of the material: lightweight material commonly provides poor sound insulation at low frequencies (Asdrubali et al., 2012). However, the sound insulation of a massive structure mainly depends on the performance of its heaviest components, such as masonry or concrete. In the case of a double wall, the presence of sound absorbing material inside the gap enables cavity resonances to be limited, thereby increasing the sound insulation of the wall.

A good acoustic solution can be achieved with massive homogeneous partitions (high-acoustic-insulation properties due to high densities) and/or insulating layers with high open porosity and incorporated air, although in order to also obtain good thermal resistance, the latter hypothesis is preferable (Bardage, 2017).

Thermal and acoustic insulation made of waste (agro, forest, etc.) has been developed very fast recently, although its research and development history is relatively short (Liu et al., 2017). As stated by Liu et al., bio-insulation deserves more research and can play an important role in the building sector in the near future.

The potential use of mussel shell as an aggregate in concrete and coating was already studied (Martínez-García et al., 2019a, 2019b, 2018, 2017) and an environmental assessment of mussel shell used in different possible building applications was also carried out (Bordello-Malde et al., 2016, 2015). This work studies the thermo-acoustic performance of mussel shell as a loose-fill material and defines specific construction solutions and construction procedures to enable its use by engineering as a thermal and acoustic insulation material. The objective is to demonstrate that mussel shell particles displays thermal and acoustic properties similar to those of conventional insulation material.

2 Material characterization

Mussel shell is a by-product of the cannery industry. In the canneries, the mussel (flesh and shell) is boiled in water, and eviscerated and mechanically separated from the byssus. The valuable result of the process is the meat of the shellfish which is canned and commercialised. As a by-product, this industry produces mainly the mussel shell and a small quantity of byssus (Figure 1).

In this research the authors analyse the possible use of the whole mussel shell (S) (Figure 2a). These shells have been heat processed to make them bacteria free (salmonella being the main concern). This heat treatment consists of heating shells at 135 ºC for 32 minutes, and in this manner complying with the European regulations for poultry feeding (European Parliament and Council, 2009) as this is one of the actual uses of the material. This procedure ensures the disinfection of the aggregate and warrants its safe handling and storage. Different size mussel fractions are obtained after this treatment, such as mussel shell gravel (MG) (Figure 2b) and, after crushing, coarse mussel shell sand (CMS) (Figure 2c) and fine mussel sand (FMS) (the latter was not used in this work). In order to develop a bio-based insulation material, three different mussel shell fractions were analysed in this work: whole mussel shell, mussel shell gravel and coarse mussel shell sand (Figure 1).

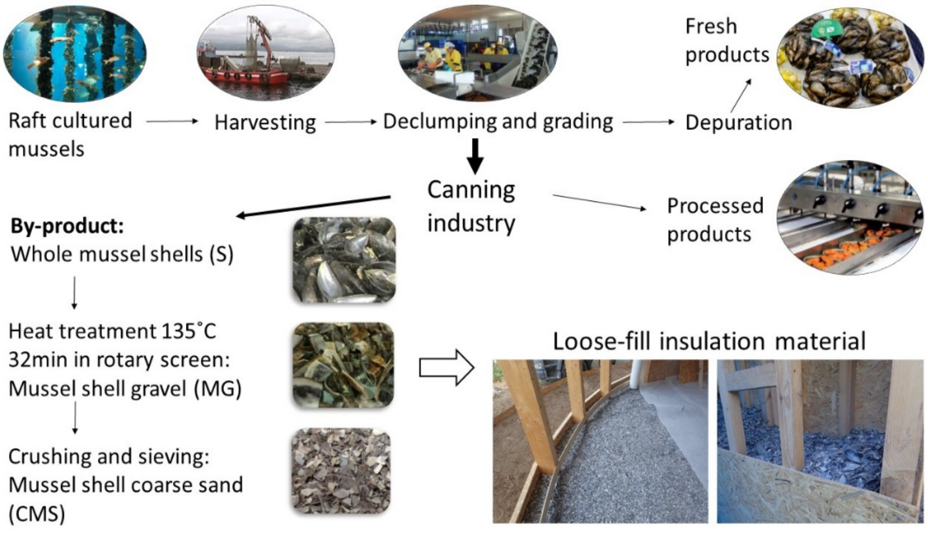

Figure 1. Process to obtain mussel shell insulation material.

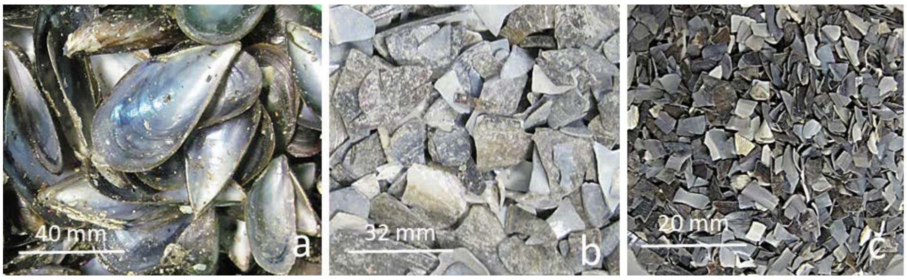

Figure 2. Mussel shell used: a) whole mussel shell (S), b) mussel shell gravel (MG), c) coarse mussel shell sand (CMS).

2.1 Physicochemical properties

Most marine shells, specifically mussel shells, are mainly composed of calcium carbonate (Martínez-García et al., 2017). With this composition, all aggregates studied in this research (gravel, sand and whole shell) show particle densities between 2.62 and 2.65 kg/l (Table 1). This density is similar to that of conventional aggregate. Regarding bulk density, this varies substantially depending on the aggregate fraction. The lower value is given by the whole mussel shell and is only 270 kg/m^3. Once the shells are crushed the voids are reduced, and so the bulk density increases with the level of crushing. The MG and the CMS show density values of 680 kg/m^3 and 1,090 kg/m^3. Water absorption of mussel shell aggregate is around 2.3 %, which is similar to the absorption of conventional aggregates.

Mussel shell aggregate mainly consists of $CaCO_3$ with a content exceeding 95 %, the rest consists of SiO_2, $MgCO_3$, Na_2CO_3 and SO_3. The calcium carbonate appears in different polymorph structures, primarily as calcite with a secondary presence of both aragonite and vaterite. This agrees with the results of other authors who analysed different types of bivalve marine shells like mussels, cockles and oysters (Adewuyi and Adegoke, 2008; Kelley, 2009).

Figure 3 shows different SEM images obtained using a JEOL JSM 6400 microscope. The images show the main material layers, with the thickest layer being the prismatic layer, which in the case of the mussels measures ~ 400 μm. This is the main resistant structure of the shell and provides the shellfish with protection. It can be classified as biomineralised $CaCO_3$ and displays a structure of prismatic calcite crystals. During the shellfish's growth, its shell needs to enlarge to provide it with space. This enlargement is made possible by growing additional crystals on top of the previous layer of prismatic calcite crystals. Further details on the physicochemical properties and microstructure of the bivalve shells are described in the work of Martinez-García et al. (Martínez-García et al., 2017).

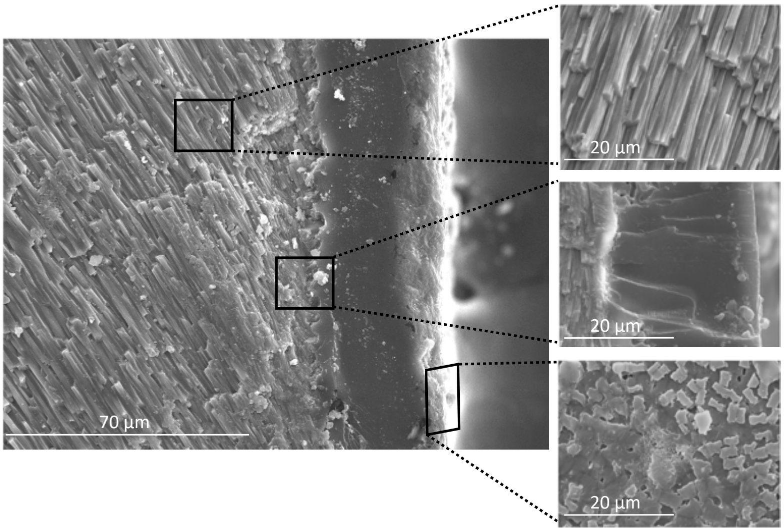

Figure 3. Shell structure (SEM images).

Table 1. Properties of mussel shell material.

	Whole mussel shell (S)	Mussel shell gravel (MG)	Coarse mussel shell sand (CMS)
Bulk density (kg/m^3) (AENOR, 1999)	270	680	1,090
Particle density (kg/l) (AENOR, 2006a)	2.62	2.62	2.65
Water Absorption (%) (AENOR, 2006a)	2.17	2.17	2.56

2.2 Classification and particle size distribution

The whole mussel shell (S) was classified according to UNE-EN 933-11 (AENOR, 2009) and the constituents of the shells were obtained as a result. In this test, a sample of 25 kg was separated and all particles were measured. It was concluded that it is mainly composed of shells with two joined valves or only one valve (Figure 4 and Figure 5). However, broken shells, clamshell bonded to the mussel shell, shells from other molluscs (cockles, oysters, clams or broken scallops) and a very small amount of byssus also appear as a part of this material. Mussel shell gravel (MG) was also classified and discovered to mainly consist of one valve of broken mussel shells (Figure 4 and Figure 5). In addition, a small amount of the mussel shell gravel material included limpets, broken white shells and byssus.

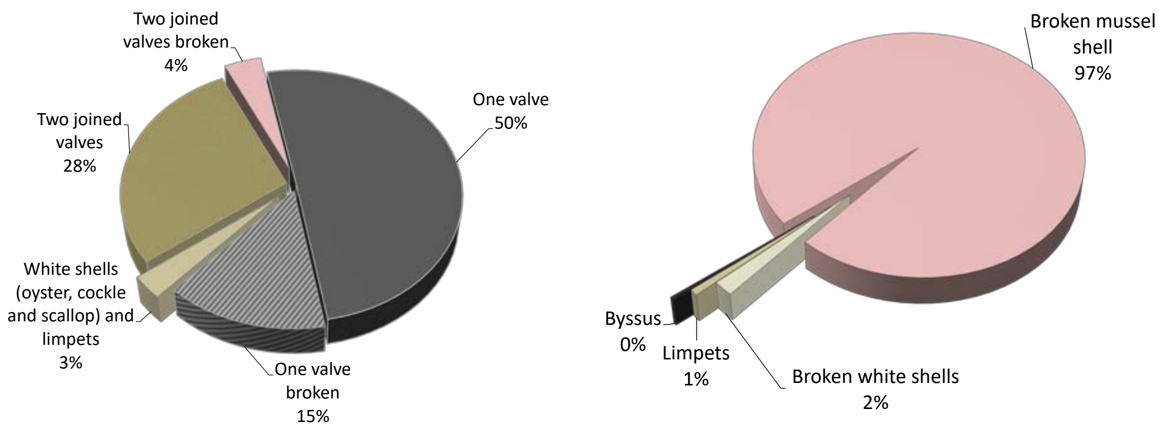

Figure 4. Composition (%) of whole mussel shell (S) (left) and mussel shell gravel (MG) (right).

Figure 5. Constituents of whole mussel shell (S) and mussel shell gravel (MG).

To characterise the shell size, direct measurements with a calliper were taken of a sample of 75 particles of the whole shell (Figure 6). The whole mussel shell length ranged from 50 to 90 mm and the width from 20 to 40 mm, with mean values of 76 mm and 37.

The next test was a sieve analysis of mussel shell aggregate (Figure 7). The grain size distribution is an important property of any granular material, however, in this case, the results are affected by the flaky shape of the particles and this limitation should be considered. After different sieving times were tested, a sieving time of 7 minutes was chosen to guarantee the smallest deviation in the particle size distribution.

The whole mussel shell presented a fineness modulus of 10.72 and particle size distribution between 20-31.5 mm. Mussel gravel showed a fineness modulus of 5.38 and particle size distribution between 4-16 mm. Finally, the mussel shell coarse sand exhibited a fineness modulus of 1.90 and particle size distribution between 0-4 mm.

The mussel shell's morphology is clearly planar, so it was decided to study the flakiness of the material. The result of flakiness index was, as expected, very high, with a total of 99 % of flaky material (Figure 8).

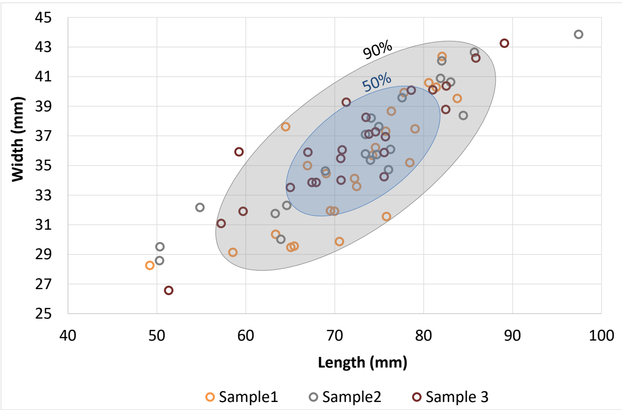

Figure 6. Whole mussel shell (S) size, length and width distribution, including 50 % and 90 % ellipsoid.

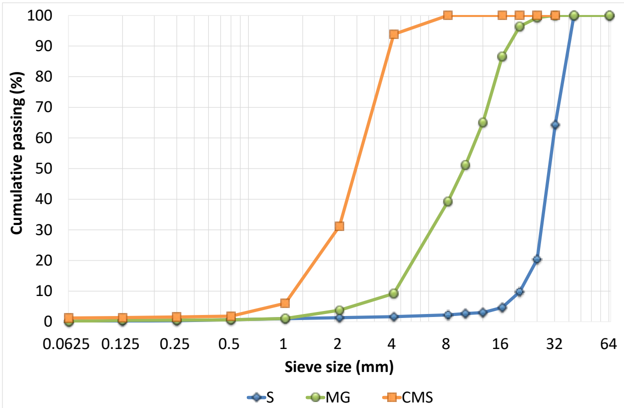

Figure 7. Mussel shell particle size distribution (whole shell, gravel and sand).

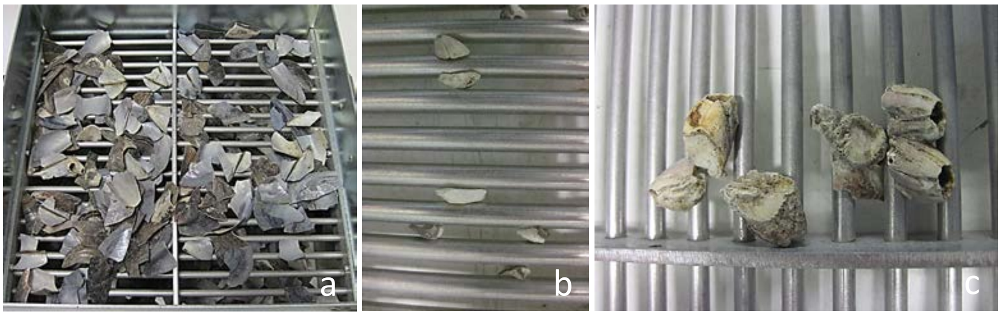

Figure 8. Flakiness index test: a) mussel shell gravel before testing; b) and c) retained material after testing.

2.1 Leaching

One of the significant parameters of any recyclable by-product is its leachability, which indicates if the waste is potentially dangerous to the environment. The most common way to asses this, is to study the materials' ability to be dissolved in water and so pollutes it. This approach is described in the European regulation that establishes the classification of waste according to its potential danger to surface water. Many substances that do not harm nature by themselves can produce substantial damage when they come into contact with water.

The Council Decision 2003/33/EC (Council of the European Union and 2003/33/EC, 2003) defines three types of waste with incremental levels of danger: inert, non-hazardous and hazardous. In this context, the mussel shells comply with the leaching limits of inert waste. However, the gravel and the sand fall into the second category: non-hazardous waste. Only one parameter exceeds the inert classification limit, which is the chloride content of both the gravel and coarse sand. This is due to the increase in specific surface area produced during crushing. Detailed information and further analysis of leaching results of mussel shell aggregate, also including fine sand, can be seen in the work of Martínez-García et al. (2017).

3 Test methods

To know the main characteristics of mussel shell aggregates to be used as loose fill material in a building solution, different tests were carried out. As mussel shell is an unconventional material, it was necessary to adapt standard tests and, in some cases, to design specific procedures and prototypes. Compaction procedure, thermal conductivity and sound reduction tests were carried out.

3.1 Compaction tests

In order to use mussel shell as a loose-fill material and avoid future settling, it is necessary to know its compaction behaviour and establish the accurate bulk density that must be achieved. Different tests were carried out to analyse the following: vibration compaction tests, specifically designed in this work, and static loading test according to NLT 351/74.

The vibration compaction test was designed to determine the bulk density that must be achieved to prevent future settling. This test simulates long-term ageing, i.e. it simulates the expected vibrations that buildings experience throughout their lifespan. It is then considered that in order to use mussel shell as an insulating material, it must be compacted up to the maximum density achieved with this test. In this manner, settlement of the loose material will be prevented, in order to avoid undesirable thermal or phonic bridges in buildings.

In this test, mussel shell aggregate is introduced with no compaction system, into a transparent methacrylate tube with internal dimensions of 190 mm in diameter and 510 mm in height. Then, the tube is introduced into the blind sieve and a cover is placed on top to prevent any material loss. The tube is set on the sieve shaker for 30, 60, 120, 180 and 240 seconds, with the material height and settling measured at each time (Figure 9). The compaction degree is then calculated, by expressing the final volume as a percentage of the initial volume $\frac{(Vf - Vi)}{Vf}$.

Furthermore, standard NLT 351/74 describes the procedure to determine the coefficient of friability of aggregates under static loading, providing information about the abrasion resistance of the aggregates. This test was used to analyse the expected compaction behaviour of mussel shell as a loose-fill material. Furthermore, the loading capacity of the material and the impact that loading has on its particle size distribution were also analysed.

In this test, a sample of mussel shell aggregate is weighed (the weight of the sample depends on the aggregate's maximum size) and placed, without any compaction system, into a metal mould of 153 mm in diameter and 125 mm in height. An increasing load was applied to the sample and the volume reduction was measured. The loads applied were 100, 200, 500 and 2,000 kg, which led to compressive stresses of 5.5, 11, 27.5 and 110 MPa. The particle size distribution of the aggregate was measured at the beginning and end of each loading process. The results provide information about the maximum loads that can be applied during the compaction process and how they affect the grading sizes of mussel shells.

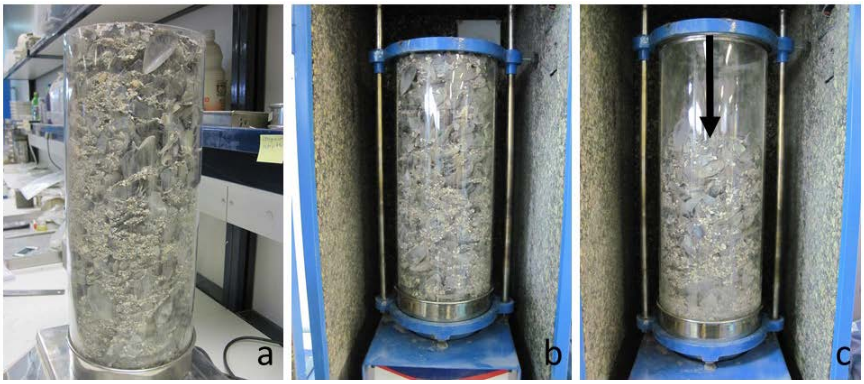

Figure 9. Vibration compaction test: a) sample inside the tube; b) sample at the sieve shaker; c) sample after shaking for 1 minute.

With the most suitable material, a prototype was made and tested combining both the compaction and vibration tests, in order to confirm the previous results. The methacrylate tube (previous one) and an almost-full-scale wall were used to characterise the prototype.

The tube was filled with mussel shells in different layers with an average thickness of 30 cm, hand compacted with a wooden rammer until the compacted density obtained in the previous test was reached. The tube was set on the sieve shaker for at least 120 seconds, and then the settling was measured.

The wall prototype was made with wooden framework consisting of pieces of a wooden stud of 70 mm x70 mm with a total length of 2.45 m. This structure was covered with wooden boards (OSB) of 19 mm. The prototype was reinforced with horizontal wooden pieces of 70 mm x 40 mm placed every 50 cm. A retractable door was placed at the bottom of the wall to remove the material once the test was finished (Figure 10).

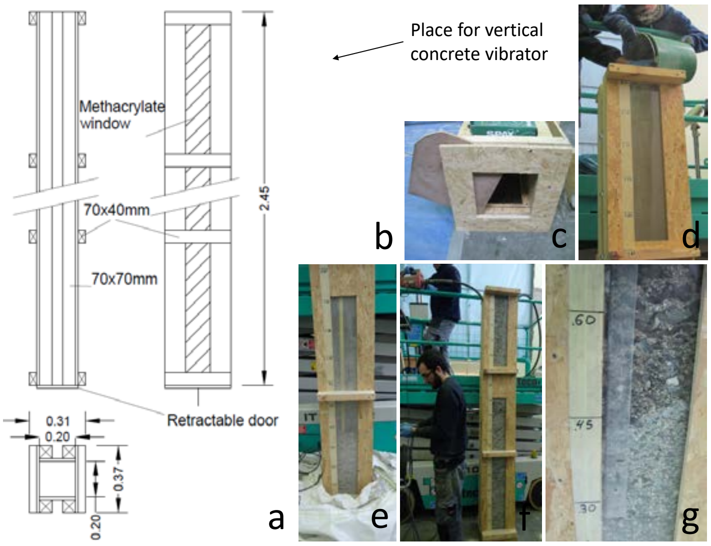

Figure 10. Wall prototype: a) wall prototype plans; b) prototype built; c) bottom retractable door; d) filling process; e) height measurement; f) shaking with vibrators; g) material after shaking.

The prototype was filled in four layers with an average thickness of 70 cm each, and hand compacted with a big wooden rammer until the compacted density obtained in the previous test was achieved. After the wall was filled, vibration was applied using two needle vibrators (50 Hz) placed on the outside of the wall. The vibrators were activated for 60 seconds to confirm that there was no settling.

3.2 Thermal conductivity test

In this work, the thermal conductivity was measured according to UNE-EN 12667 (AENOR, 2002) (guarded hot plate and heat flow meter methods) , the standard recommended for materials with medium and high thermal resistance. The EN 12667 standard recommends a range of 10 K to 50 K in the temperature difference between the hot and the cold face. A difference of 20 °C was established in the mussel shell test as this value ensures that the heat flow that crosses the sample is within the measurement range of the equipment used. In addition, to fix the mean temperature of the sample, the UNE-EN 14063-1 (AENOR, 2006b) was followed. This standard states that the reference mean temperature shall be 10 °C. The equipment used for the test is a device based on the HFM 436/6/1 Lambda heat flow meter method. The control and data acquisition software used was Q-Lab 2. The maximum dimension of the samples to be tested was 60 cm x 60 cm x 10 cm. The measurement was performed on a sample central area of 30 cm x 30 cm.

As aforementioned, mussel shell is a granular material, a non-homogeneous mixture consisting of a random distribution of shells and intermediate air cavities. The mussel shells were confined in boxes made of plain laminated plywood boards, which were fixed with glue and nails. Characteristics of the sample boxes created and mussel aggregates tested for the thermal conductivity test are shown in Table 2 and Figure 11. The bulk density and porosity of each sample were determined according to UNE-EN 1097-3 (AENOR, 1999)**.**

Table 2. Samples characteristics for thermal conductivity test.

Sample	Box dimensions (mm)	Bulk density (kg/m³) (AENOR, 1999)	Porosity (%)(AENOR, 1999)
S	599x598x108	281	89
MG	599x600x108	684	74
CMS	600x600x65	1,205	54

Figure 11. Boxes for thermal test: a) entire shell, b) mussel shell gravel, c) coarse mussel shell sand, d) box in the heat flow meter device.

The hot plate (T_H) was in contact with the mussel shells, and the cold plate (T_C) was in contact with the bottom wooden board (Figure 12). During the test, five points of heat flow were distributed over the surface of each type of mussel shell sample.

Using this system, the thermal resistance is the sum of the thermal resistance of the different parts of the sandwich system design (mussel shell + bottom wooden board). The thermal resistance of the loose-fill material (i.e. mussel shell) was determined by deducting the thermal resistance of the wooden board from the total thermal resistance of the sandwich system (Figure 12).

Thermal resistance of the wooden board was previously determined according to ASTM C-1114 (ASTM International, 2019) using specimens of 70 mm x 110 mm x 7 mm (Figure 13). The value obtained was λ_b = 0.12 ± 0.01 W/(m.K).

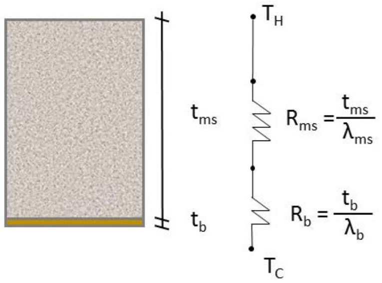

Figure 12. Thermal circuit with two thermal resistances: the inhomogeneous loose-fill material (mussel shell) and the laminated wood board.

Figure 13. Wooden boards (70 mm x 110 mm x 7 mm) tested according to ASTM C-1114.

3.3 Sound reduction test

Airborne sound insulation was measured in a reverberation room prepared for testing the sound insulation of glazing systems (Figure 14). The test was carried out according to UNE-EN ISO 10140-2 (AENOR, 2010) measuring the airborne sound insulation with sound reduction index, R (in decibels).

Figure 14. Reverberation room to carry out sound reduction tests.

The sound reduction index defines the ability of a structure (wall, roof, window, etc.) to prevent the passage of sound through itself and is expressed in dB (Asdrubali et al., 2015). This index is calculated by measuring the time- and space-average of the sound pressure level in both rooms separated by the partition being tested.

The tested sample is placed in a gap at the centre of the partition. A sound pressure level is generated in one of the rooms, named the source room, to be received in the other room named the receiving room. According to standard (UNE-EN ISO 10140 (AENOR, 2011a)) two different corrections must be considered:

- Correction due to flank sound transmission (UNE-EN ISO 10140-2 (AENOR, 2010)): no correction due to flank sound transmission is required, if at any frequency, the partition displays a sound reduction index R (calculated following the standard procedure given in ISO 717-1 (AENOR, 2013)) that is 6 dB higher than the sound reduction obtained using the tested sample.
- Correction due to background noise (UNE-EN ISO 10140-4 (AENOR, 2011b)): a correction must be made if there is background noise that is 15 dB higher than the sound generated in the room during the test.

The sound reduction index, R (dB), is calculated using the following equation (1):

$$R = L_1 - L_2 + 10\log\frac{S}{A} \quad (2)$$

where L_1 is the sound pressure level in the source room, L_2 is the sound pressure level in the receiving room, S is the surface area of the element separating the rooms (m^2) and A is the equivalent absorption area in the receiving room (m^2). The latter is calculated using the following equation:

$$A = 0.161\frac{V}{T} \quad (3)$$

where V is the volume of the receiving room (m^3) and T is the reverberation time in the receiving room (sec). In case of background noise exceeding 15 dB, a correction must be applied to the L_2 measurements.

According to the standards ISO 10140-5 (AENOR, 2011c) and ISO 10140-4 (AENOR, 2011b), the following measurement equipment was used:

- dual channel symphony sound level meter marked 01 dB Metravib model 5470;
- Rion brand calibrator with serial number 35173579 model NC-74;
- GRAS 40 AF microphone with serial number 77375;
- GRAS 40 AF microphone with serial number 77374;
- computer program dB01;
- dodecahedral speaker.

In this work, the volume of the source and reception rooms were 50.2 and 50.5 m^3. The partition consisted of a double leaf full brick wall (150 mm each leaf) containing a 4 mm thick insulation board of mineral wool in its cavity. This partition was previously designed to take the correction due to flank sound transmission into account where necessary.

A prototype partition wall was designed to carry out this test. The prototype (Figure 15) consisted of a big box made of a wooden frame and OSB board with a density of 650 kg/m^3 and thickness of 15 mm. The external dimensions of the box were 1,490 mm x 1,240 mm x 120 mm. The box was filled with mussel shell gravel with an average density of 676 kg/m^3. The thickness of the loose-fill material was 90 mm. The prototype was installed inside a test bench like a window, and it was sealed to the bench with a silicone mastic, which was let dry for 24 hours just before the test was done. The environmental conditions for the test were as follows:

- pressure = 100,300±120 Pa;
- humidity = 39±10 %;
- emission room temperature = 18.4±3 ˚C;
- reception room temperature = 18.1±3 ˚C.

Figure 15. Sound reduction test. Left: mussel shell wall sample before being placed. Right: sample placed into the test bench.

The sound pressure level and reverberation time values were recorded in 1/3 octave bands, and in order to obtain a single number, the weighted sound reduction index R_w was calculated following the standard procedure given in ISO 717-1 (AENOR, 2013). The overall acoustic reduction index (R_A, R_{Atr}) for the exterior noise of a building element, is the overall assessment of the sound reduction index in dB using the spectrum adaptation terms, C and C_{tr}, for medium-high frequencies and medium-low frequencies.

$$R_A = R_W + C \tag{4}$$

$$R_{A,tr} = R_W + C_{tr} \tag{5}$$

4 Results and discussion

The results obtained with the test methods are described and discussed. Firstly, settling and strength after compaction are analysed. Thermal and acoustic performance are studied comparing mussel shells with other insulation materials described in the literature.

4.1 Settling and strength

Results obtained with the vibration compaction test (Figure 16) show that the three materials (S, MG and CMS) display quick settling in the first 60 seconds of shaking. From 60 to 120 seconds all materials exhibit slight settling. Lastly, from 120 seconds MG and CMS reach a stable density and compaction degree that does not vary from then onwards (35 % and 15 %). However, S still presents settling up to 240 seconds, at which time it reaches a compaction degree of 30 %. It can be seen that despite S having less bulk density than MG, the compaction degree obtained with the latter is 5 % higher than that obtained with the former.

Mussel gravel displays the best behaviour (less bulk density than CMS and higher compaction degree than S) for use as a loose-fill material. With these results the CMS is ruled out for use as thermal and acoustic material due to its high density. It was decided to carry out the static loading test with both mussel shell and mussel shell gravel.

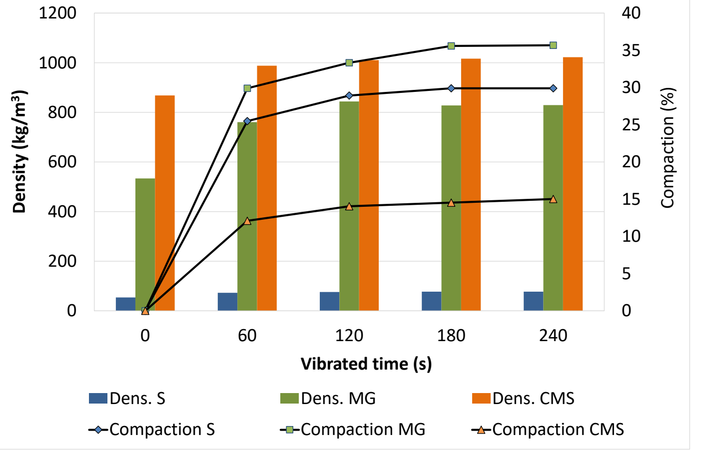

Figure 16. The density and compacting degree of mussel shell aggregates (whole, gravel and coarse sand) after different shaking times.

Figure 17 and Figure 18 show the particle size distribution of the whole mussel shell (S) and mussel shell gravel (MG) after different loading processes (which led to different maximum compressive stresses). Due to friability of the mussel shell aggregate, all of the stresses produced change with its particle size distribution. Differences between the particle size distribution at the beginning (Initial) and end of the test are more noticeable with the whole mussel shell than the mussel shell gravel. The fineness modulus was also calculated. This modulus is shown in Figure 19, and it can be seen that both materials display the same fineness modulus when the stress is 110 MPa.

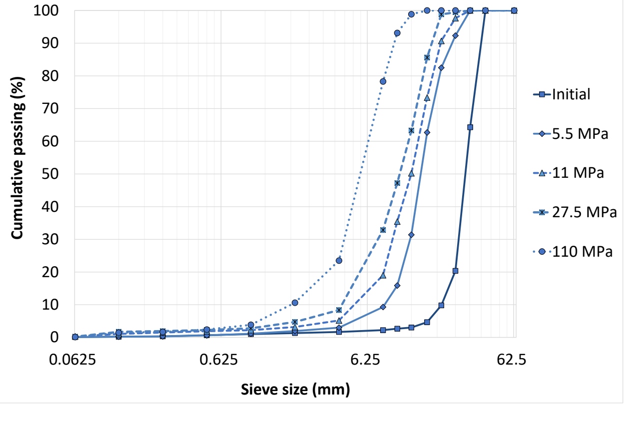

Figure 17. Particle size distribution of whole mussel shell (S) under different loads.

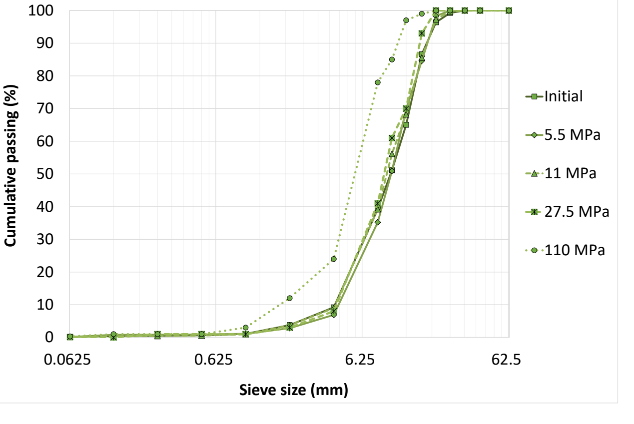

Figure 18. Particle size distribution of mussel shell gravel (MG) under different loads.

The volume before and after loading was also measured and used to calculate the bulk density (Figure 19). The results reveal that the density of S, after being loaded at 5.5 MPa, is twice its initial value and increases by more than 300 % when 110 MPa is applied. Regarding mussel gravel, density values are not affected until 27.5 MPa is reached.

The results provide information about the maximum loads that can be applied during the compaction process and the grading sizes of mussel shells that are least affected by loading. According to these results, it can be concluded that mussel shell gravel is able to maintain its particle size distribution up to 11 MPa; so this material can be suitably used as a loose-fill material in foundations. However, whole mussel shell is not reliable as structural material for foundations.

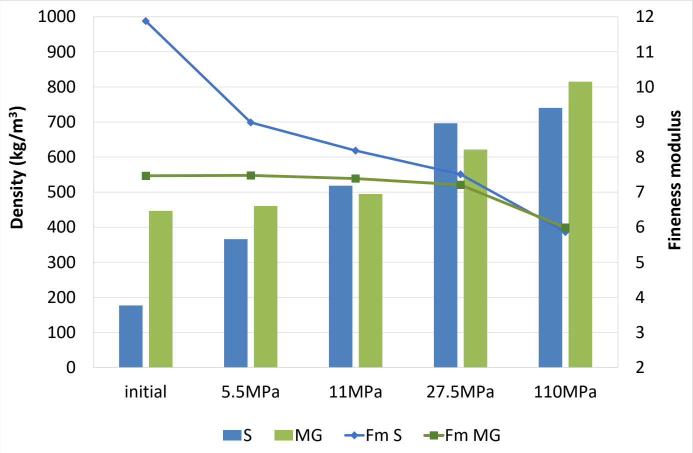

Figure 19. Fineness modulus and bulk density under different loads.

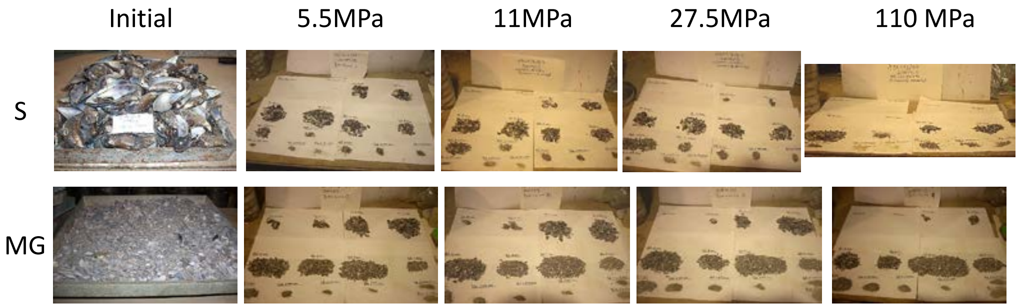

Figure 20. Mussel shell (whole and gravel) fractions after loading.

Analysing these results, MG was chosen to carry out the prototype tests. In both prototypes (tube and wall) mechanical compaction was applied to obtain the target compaction degree of 35 % (obtained in the previous test). After the mechanical compaction, both prototypes were vibrated according to the test protocols described in section 3.1 and the settlements were measured, obtaining values of less than 3 %. This value is low enough to confirm that mussel shell gravel compacted up to density values of around 1,100 kg/m^3 (compaction degree between 30 % and 40 %), can be applied as loose-fill material while ensuring no settlement. These results are shown in Table 3.

Table 3. Settlement obtained with the prototypes using mussel shell gravel.

Sample	Average compaction degree (%)	Final bulk density (kg/m^3)	Settlement after shaking/vibration (%)
MG Tube sample 1	34 ±0.07	1,115.6	2.94
MG Tube sample 2	40 ±0.003	1,258.3	0.78
MG Tube sample 3	31 ±0.02	1,072.9	1.96
MG 20x20 Wall	36 ±1.6	1,024.4	2.12

4.2 Thermal behaviour

The total thermal conductivity value obtained of mussel shell as loose-fill material is shown in Table 4. The value obtain for whole mussel shell is in agreement with the value obtained by Aagaard and Moller (Aagaard and Moller, 2007) with similar density.

Table 4. Thermal conductivity of mussel shell as loose-fill material.

Material	λ [W/(m.K)]
Whole mussel shell (S)	0.12±0.01
Mussel shell gravel (MG)	0.15±0.01
Coarse mussel shell sand (CMS)	0.20±0.01

Most authors agree with the fact that a material can be considered thermal insulation if its conductivity is less than 0.1 W/(m.K) (Mati-Baouche et al., 2014). It can be stated that mussel shell thermal conductivity is close to this value. Anyway, mussel shell thermal conductivity is considerably lower than the thermal conductivity of conventional building materials as hollow bricks (0.45 W/(m.K); 1,500 kg/m^3) (Fioretti and Principi, 2014) or dry plain concrete (1.37-2.77 W/(m.K) (Khan, 2002)).

Porosity is one of the main properties affecting a material's thermal behaviour. Intra-particle porosity is the porosity of the particle and inter-particle porosity represents the volume of voids between the particles (related to bulk density). In an aggregate with a low real density, that is, with a very high intra-particle pores volume both inter-particle and intra-particle pores are influencing its thermal behaviour. However, when the material presents a high real density (that is a material with very low intra-particle pores volume; which is the case of mussel shell), only the inter-particle pores are affecting its thermal behaviour. As mussel shell microstructure does not have intra-particle air voids, its thermal behaviour is mainly related to the air between particles. Figure 21 and Figure 22 show the thermal conductivity of mussel shell material versus inter-particle porosity and bulk density.

According to the results obtained in section 4.1, when mussel shell is compacted up to density values of around 1100 kg/m^3 (compaction degree that ensures no settlement) the thermal conductivity will be of about 0.175 W/(m.K).

All insulation materials serve the same purpose: to reduce the rate of heat release/gain through an enclosed space. Some authors carry out the classification of insulation material according to its form and composition (Aditya et al., 2017). A comparative analysis of mussel shell with other insulation material is shown in Figure 22. According to this figure, six groups are selected for comparison.

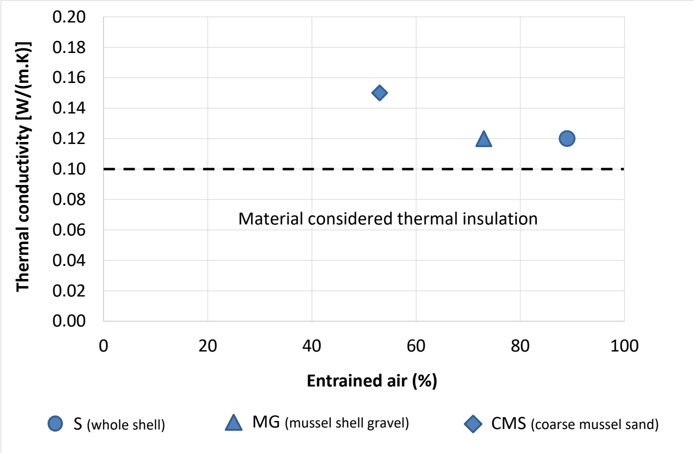

Figure 21. Thermal conductivity vs entrained air (porosity) of the loose-fill mussel shell material.

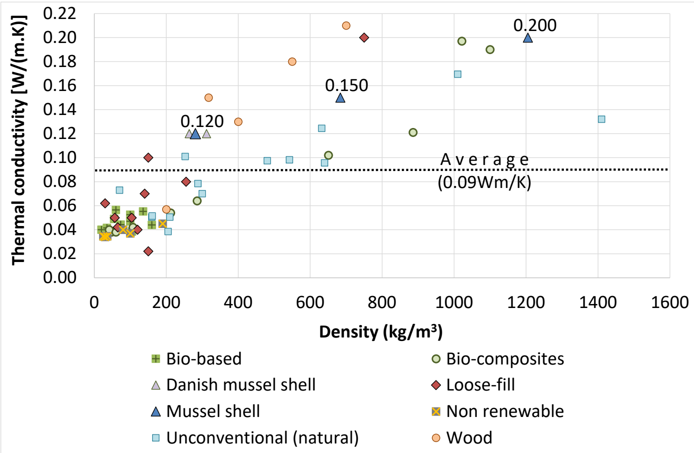

Figure 22. Comparative Thermal conductivity vs bulk density of insulation material and mussel shells.

The first group corresponds to granular loose-fill material with an open cell structure (Aditya et al., 2017) that can be blown-in or poured-in to the cavities of walls, roofs or floors. The following materials are included in this group: rock wool (loose) (Rockwool, 2017), cellulose (ground-up waste paper) (Nicolajsen, 2005), expanded perlite (natural glassy volcanic rock) (Aditya et al., 2017; Al-Ajlan, 2006), expanded vermiculite (phyllosilicate heated) (Schiavoni et al., 2016), expanded clay (Al-Ajlan, 2006), expanded cork granules (Al-Homoud, 2005; Nicolajsen, 2005), and cellular/expanded or foam glass (Hill et al., 2018).

Conventional non-renewable foam material or that commercialised as blankets are included in the second group. This is the most used insulation material in the construction sector. This group includes low-density fibreboard, glass wool (Rockwool, 2017), stone wool (Karamanos et al., 2008), expanded polystyrene (EPS) (Lakatos and Kalmár, 2013), extruded polystyrene (XPS) (Vo et al., 2011) and polyurethane (Asdrubali et al., 2012).

Bio-based material commercialised as blankets, boards or blocks is included in the third group. All of these are environmentally friendly and commercially demanded by the sustainable construction industry. Sheep's wool (Zach et al., 2012), coconut fibres (Manohar et al., 2006), cotton (Binici et al., 2013), wood fibres (Steico, 2020), hemp fibres (Cannabric, 2009), hemp shiv (Laborel-Préneron et al., 2018), flax (Kymäläinen and Sjöberg, 2008), straw bale (Chaussinand et al., 2015) and kenaf (Korjenic et al., 2011). Within the group of bio-based materials, two other groups or divisions can be made: bio-composites and wood. As bio-composites are included: compressed earth blocks (CEB) with lime and hemp fibres (Cannabric, 2009), rice husk-earth composite (Antunes et al., 2019) and different composites studied by Palumbo et al. (2016) (corn pith with sodium alginate, barley straw with corn starch, hemp fibre with adhesive and additives, hemp hurds with lime, wood wool with adhesive and additives, wood fibre with additives). Five types of wood with different densities: oak, spruce, pine, eucalyptus and balsa wood (CTE web, 2007), are included.

Natural unconventional insulation materials that are not or scarcely commercialised are included in the fourth group. This material is analysed in the work of Asdrubali et al.(2015). Material included in this group is as follows: banana and polypropylene fibre, bagasse, corncob, cotton stalks, date palm, durian, oil palm, pecan, pineapple leaves, reeds, rice, sansevieria fibre and sunflower.

Comparing the thermal behaviour of mussel shells with these materials, it can be seen that expanded clay (loose-fill) presents a similar thermal conductivity (in the range of 0.08 (255kg/m^3) - 0.20 (750 kg/m^3) W/(m.K)) to that of mussel shells.

Wood species display the same thermal behaviour as mussel shell. However, the thermal conductivity of wood is low due to its porous structure and several air-filled cell lumens (Jones and Brischke, 2017). Commercialised compressed earth blocks with hemp fibres (Cannabric, 2009) and mussel shell have a similar conductivity behaviour. Hemp fibres ith a lambda value of 0.048 W/(m.K) are responsible for a great reduction in the thermal conductivity of a common CEB (0.77 W/(m.K); 1.8 kg/m^3) (Abdullah et al., 2010).

Regarding natural unconventional materials, many display similar conductivity values to those obtained with mussel shell. For instance, corncob particleboards can be compared with mussel shell. Corncob is waste that comes from the corn processing industry. Corncob particleboards are made from ground corncobs agglomerated with wood glue, and their thermal conductivity is estimated by different authors (Asdrubali et al., 2015), where the best value reached was 0.101 W/(m.K). The thermal conductivity of this material is based on its microstructure.

Particleboards made of durian peel and coconut coir fibres also display thermal characteristics similar to those of mussel shell. Durian peel and coconut coir fibre particleboards with different densities and binders were studied by Khedari et al.(2003), finding λ values between 0.064-0.185 W/(m.K).

In the study of Evon et al. (2014) thermal insulation fibreboards were created by thermo-pressing cake generated by the biorefinery of whole sunflower plants in a twin-screw extruder. These fibreboards displayed thermal conductivity values between 0.0885 and 0.110 W/(m.K), comparable to those measured using the whole mussel shell.

Lastly, a banana-polypropylene fibre composite studied by Paul et. Al. (2008), with λ values between 0.157-0.182 W/(m.K), can also be compared with mussel shell. In most cases, the thermal insulation of these composites is based on their inter-particle porosity.

In conclusion, the thermal behaviour of mussel shell is in the range of some natural unconventional bio-based and loose-fill material, and displays a behaviour that is suitable for application as insulation.

4.3 Acoustic properties

Results of acoustic properties are shown in Table 5. Lastly, to carry out this test according to the previous results, mussel shell gravel was selected as the loose-fill material. According to these results, it was not necessary to make any corrections due to flank sound transmission or background noise.

According to the recommendations of the Spanish Technical Building Code (CTE-DB-HR, 2009) shown in Table 6, the results obtained show that the solution is suitable for use as a partition wall, façade enclosure (for quiet areas) or/and dividing wall between different buildings.

Table 5. Sound reduction index of mussel shell gravel.

Description	Value (dB)
Global index of sound reduction R_w (C,C_{tr})	42 (-1;-3)
Sound reduction index of airborne sound (medium-high frequencies) R_A	41
Sound reduction index of airborne sound (medium-low frequencies) R_{Atr}	39

Table 6. Recommended sound insulation index (R_A) according to the Spanish standard (CTE-DB-HR, 2009).

Adjoining place	Façades (1)	Partition wall	Party wall (2)	Common zones	Industrial/activity facilities
Acoustic requirement	≥ 30 - 47 dB	≥33 dB	≥ 40 dB ≥ 50 dB	≥ 50 dB	≥ 55 dB

(1) Minimum and maximum data reflected in CTE-DB-HR (2009), depending on the external noise, type of building and outside noise levels during the day.

(2) ≥ 40 dB for each of the building´s enclosures ≥ 50 dB for both as a set.

Figure 23 shows the sound behaviour of different materials that are commonly used as acoustic insulation solutions, both natural and conventional. These materials have been selected as they were used in solutions with a similar width to the mussel shell sample designed in this work. According to the work of Desarnaulds et al. (Desarnaulds et al., 2005), the mussel shell solution can be compared to a light weight double wall (inner and outer gypsum board of 13 mm and a wooden frame). This wall is filled with 100 mm of different insulation material such as glass wool (batts, 15 kg/m^3), flax (batts, 35 kg/m^3), cellulose (batts, 25 kg/m^3) and cellulose (loose-fill 50 kg/m^3). The sound reduction index (Rw) obtained with this solution was 40 dB for the first three insulation materials and 41 dB for cellulose used as loose-fill material.

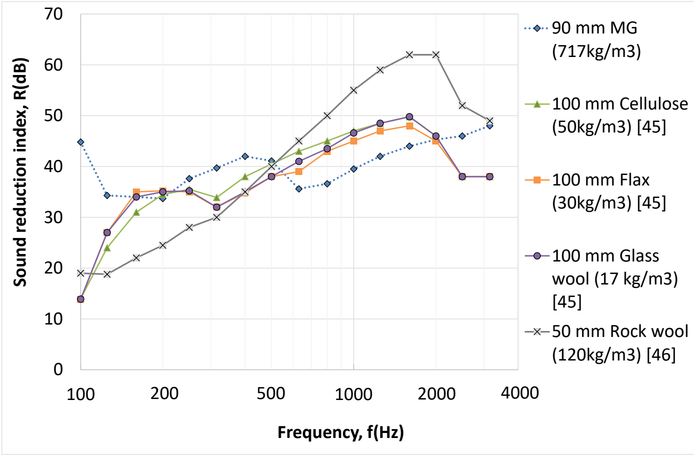

Figure 23. Sound reduction index of mussel shell gravel and different conventional and natural insulation materials.

Uris et al. (1999) studied the influence of different rock wool densities on the acoustic properties of a partition wall. The highest rock wool density solution was selected for comparison with the mussel shell results. Uris et al. (1999) designed a 76 mm thick double-leaf partition consisting of a 50 mm frame of steel studs and single layer on each side of the gypsum board. The space between the gypsum boards was filled with rock wool with a density of 120 kg/m^3. This solution provides a sound reduction index (Rw) of 41 dB.

Mussel shell shows better acoustic behaviour (sound reduction) than almost all of the materials at low frequencies (100 to 500 Hz). The mussel shell values obtained at frequencies between 500 and 2,000 Hz are below those of cellulose, flax or glass wool. These differences reach 26-40 % when rock wool is used as a comparison. At high frequencies (> 2,000 Hz) mussel shell is better than cellulose, flax or glass wool.

Al low frequencies, porous materials have to be considered as a lumped element circuit being, in this case, the main factor affecting sound reduction, the resistance and mass inertance of the material (Bies and Hansen, 1980). Mussel shell presents considerably higher density values than any of the other insulation materials, which leads it to show the best acoustic behaviour at low frequencies.

5 Mussel shell as a sustainable unconventional insulation material

A comparative analysis of mussel shell with other commercial insulating materials, both natural and synthetic is carried out. The advantages and disadvantages of its use on site are evaluated from a functional point of view. The environmental evaluation of mussel shell as an insulating material is also analysed based on the work carried out by Bordello et al.(2016, 2015).

5.1 Mussel shell compared with other materials

As already shown (section 4.1 and 4.2), the behaviour of mussel shell gravel as a loose material can be compared to conventional insulation material such as expanded clay, both regarding thermal behaviour and placement technique. Nevertheless, mussel shell gravel has its advantages and disadvantages when compared to other commercial materials.

The open pore structure of some of the natural material presents a drawback due to high wettability, which means that they must be protected against biological attacks (fungi, moulds, parasites, etc.) and fire. As a result, different authors (Bardage, 2017) propose the use of hydrophobic agents to mitigate the material's degradation. However, it must be considered that the agent itself affects the thermal behaviour of the insulation material. Mussel shell (mainly consisting of calcium carbonate) is stable against biological attacks and fire, which is a significant advantage when compared with other bio-based insulation material.

Nowadays the most used insulating material is blankets, boards or rigid foams. These are easy to install in unfinished walls, floors and ceilings and can be transported efficiently due to their low density. On the other hand, loose-fill and blow-in materials are suitable for installation in enclosed existing walls, wall cavities, unfinished attic floors and hard-to-reach areas. Mussel shell material can be included in this last group, although its density and placement protocol (manual or mechanical compaction is necessary to guarantee no settlement) hinder its use in retrofitting or refurbishment.

According to the results of this work, the most implementable applications of mussel shell in building solutions are the following:

- Walls: the compacted mussel shell can be placed inside a wooden frame structure, and confined with wooden boards and coatings on both sides. This solution is flexible in width and can be carried out for envelopes and partitions.
- Roofs: it can be applied with inverted flat roof solutions, where compacted mussel shell acts as a thermal and acoustic insulation material. A wooden board waterproofed by laying EPDM type rubber sheets, can be placed over the mussel shell. A green roof can be also built using mussel shell as a finishing coat, although, in this case for aesthetic purposes.
- Floors: the compacted mussel shell can be placed directly on the soil, instead of conventional drainage material like limestone gravel. Then a concrete slab or wooden structure can be put in place, and lastly, the finishing floor is applied. When compared with limestone gravel, compacted mussel shell displays many advantages: it is more stable and comfortable to work on; it has low thermal conductivity; better acoustic behaviour and; acts as a capillary barrier (Martínez-García et al., 2019b, 2019a, 2017).

5.2 Embodied energy of mussel shell as loose-fill material

Bordello et al. (2016, 2015) carried out an assessment of the sustainability of mussel shell as an insulation material, under the framework of the project “Assessment of Galician bivalve shell in the construction sector”. This work aimed to evaluate the integral sustainability of mussel shell by considering environmental, social and economic aspects. Furthermore, the sustainability of mussel shell when used as an insulation material, was compared to that of the conventional insulation material, expanded clay.

The method used was based on the MIVES technique (Integrated Value Model for a Sustainable Evaluation), based on requirement trees, value functions and the Analytical Hierarchy Process: AHP. The latter was used as a support to establish the weights of the different parameters of the sustainability evaluation model.

A cradle-to-gate (partial assessment of the product life cycle form resource extraction –cradle- to the factory gate, i.e. before it is transported to the consumer) approach was used to assess the real environmental impact of the mussel shell, which is the most common analysis carried out by companies commercialising construction material or systems. The cradle for the mussel shell was located at the canning facilities where shells are stripped from the mussels and collected to be taken to the company that processes, prepares and sells them. The gate for the mussel shell was defined as at the facilities of the shell processing company where the new product is ready for sale.

According to the model presented by Bordello et al. (2016) the Global Sustainability Index of expanded clay was 0.35 and that of mussel gravel, 0.71. Taking into account the three considered aspects of sustainability (environmental, social and economic aspects), the mussel shell improves the conventional option in all of them. The highest improvement is reached in the economic aspect: mussel shell is an economical option and contributes to savings in waste management by canning companies. The advantage of the mussel shell in the social field is mainly due to the fact that cannery industry is a local female sector and to the fact that it promotes R+D+I projects. Regarding environmental aspect, differences are low: emissions are slightly higher for mussel shell; however, its use avoids natural resources consumption, preserves the ecosystem and promotes waste consumption.

In order to make a comparative analysis with other different thermal insulation materials, only the Embodied Energy of mussel shell calculated by Bordello et al. (2016) was considered. This Embodied Energy was calculated using a Functional Unit (FU) defined as the quantity of material necessary to ensure a thermal resistance of 1 m^2K/W for 1 m^2 of wall. With a mussel shell estimated density of 570 kg/m^3 a functional unit of 63.84 kg was obtained. According to these results, the corresponding Embodied Energy calculated for mussel shell gravel was 33.035 kJ/UF.

Figure 24 shows the embodied energy values (MJ-Eq per kg) of mussel shell gravel and of other different thermal insulation materials that were taken from the literature (Ardente et al., 2008; Bordello-Malde et al., 2016; Intini and Kühtz, 2011; Schiavoni et al., 2016; Zabalza Bribián et al., 2011). They were all assessed using cradle-to-gate life cycle analysis. It is clear that mussel shell shows very low Embodied Energy values, like other insulation material such as natural pumice, cellulose, hemp and kenaf fibre, and considerably lower values than expanded polyurethane, glass fibres, etc.

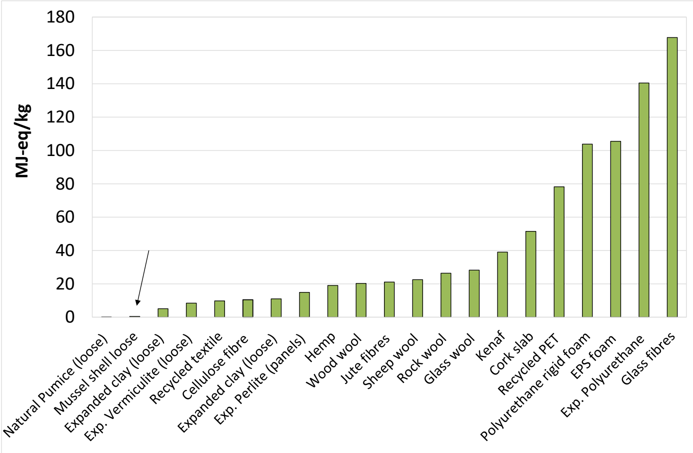

Figure 24. Embodied energy of different thermal insulation materials (cradle-to-gate approach).

6 Conclusions

This study was conducted with the aim of proving the feasibility of using mussel shell as a bio-based insulation material in sustainable building solutions. There was a special focus on the thermal and acoustic properties of mussel shell. Different mussel fractions were analysed and according to the results obtained, the following conclusions were drawn:

- To avoid long-term settling it is necessary to compact the material. Mussel shell gravel provides the best performance as, in the vibration compaction test, it displays less bulk density than coarse mussel shell sand and a higher compaction degree than whole mussel shell. Furthermore, it can maintain its particle size distribution up to 11 MPa. According to the prototype tested, no long-term settlement is expected using this material with a compaction degree of 35 %. This leads to conclude that this material is suitable for use as loose-fill material in foundations and wall or roofs cavities.
- The thermal conductivity value of mussel shell is close to that required to be considered a thermal insulation material (0.17 W/(m.K) vs 0.10 W/(m.K)). The average value of the three different samples were similar to the thermal properties of wood, which allows for concluding that it displays suitable behaviour for insulation applications.
- The acoustic characteristics of the shells demonstrate that the designed solution is suitable for use as a partition wall, façade enclosure (for quiet areas) and/or dividing wall between different buildings. Its sound reduction index is similar to conventional insulation material (cellulose, flax or glass wool, etc.). Due to its density, it shows very good acoustic behaviour at low frequencies (100 to 500 Hz).
- According to the results of this work, different mussel shell applications in building solutions were proposed. The most promising solutions were complete systems for roofs, walls and floors and one of the main advantages as a bio-based building solution is its stability against biological attacks and fire.

- Lastly, regarding environmental performance, mussel shell shows very low values of embodied energy, like other insulation materials such as natural pumice, cellulose, hemp and kenaf fibre, and considerably lower values than expanded polyurethane, glass fibres, etc.

7 Acknowledgement

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