Production and characterization of sandwich composite materials with cork agglomerate core

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Abstract

The use of composite materials, namely sandwich structures, has been increasing exponentially in the most diverse industries. This is due to the fact that, in this class of material, by combining properties from different components it is possible to achieve an optimization of the final composite properties. Thus, depending on the intended application, these materials can be lighter, stronger, or less expensive when compared to conventional materials. This work aims to develop characterization methods for the distinct components of a sandwich composite, with cork agglomerate core and woven fabric and epoxy resin skins, in order to better understand the properties of each constituent and enable to compare different materials for the same purpose in the final composite. After characterizing the manifold components, it is intended to understand its influence on the final composite stiffness. Therefore, several fibre-core combinations were developed, always using the same epoxy resin, which were subsequently submitted to a bending test. The obtained results allow us to understand the influence of each component on the final composite, as well as to identify the advantages and disadvantages of the use of each configuration.

Keywords: composite materials, sandwich structure, cork agglomerate, characterization method

1. Introduction

Materials have accompanied humanity since the beginning of its existence. Among the first materials used by man were stone, wood, bones, feathers, shells, animal skin and clay, each one serving for a specific purpose. Materials were mostly used for tools, weapons, shelter or for self-expression, and it is unmistakable that the evolution of civilizations led to the invention, development and use of increasingly elaborate materials.

Nowadays, with the strong industrial competition, there is also a tireless pursuit for high performance materials. Upon that, interest in composite materials, including sandwich structures, has steadily increased in different areas, from the aeronautical to the construction sector.

With the increase in demand, new configurations and ideas for the use of alternative materials also come up. It is in this context that cork agglomerates emerge, which, due to their high mechanical resistance to shear, low specific weight, high compressibility, good thermal and acoustic insulation and vibration suppression, are increasingly used as a core of sandwich composites.

Therefore, the need to develop methods to characterize and compare the different constituents of this type of sandwich structures arises, as well as the need to understand the influence of each component on the properties of the final composite.

In this way, this work purpose is to develop and establish methods which allow to characterize and compare each one of the constituents of the final composite (cork agglomerate, glass and carbon reinforcing fibres and epoxy resin), along with the production of four different sandwich configurations, by using several constituents materials. After that, mechanical and physical properties of configurations are analysed, in order to better understand the contribution of each component to the final characteristics of the composite.

Sandwich composites were mainly driven by the aeronautical industry, due to their high mechanical properties combined with a low specific weight. The structures of this type are, in their simplest form, composed by two parallel thin sheets of a high strength structural material (skins), separated from each other by a thicker material with a lower density (core). Since, in general, the core is fixed to the skins by an adhesive layer, as shown in Figure 1.

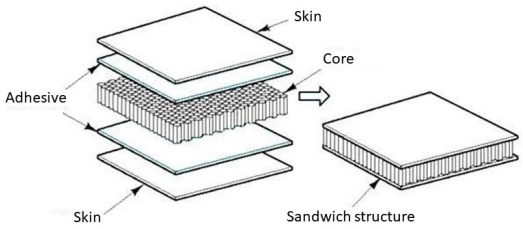


Figure 1: Sandwich structure (adapted from [2])

1.1. Sandwich composite using cork agglomerates as core

Cork is a natural cellular material that acts as a protective layer of cork oak (*Quercus suber L.*), being the outer covering of its trunk and branches [7].

Due to their peculiar mechanical, physical and chemical properties cork is used for several applications in distinct industries. For example, in the aerospace sector, incorporation of cork began with the Apollo 11 mission which, in 1969, took Man to the Moon for the first time. Since then, due its thermal isolation properties, low combustions rate and shock absorption capacity, cork has been chosen for a few applications in this sector [1].

When comparing cork agglomerates to other core material, like honeycombs or foams, they have a high damage tolerance to impact loads, exceptional damping characteristics for suppression of vibrations, and also good acoustic and thermal insulation capacities [3].

Cork is also a material of great value to the Portuguese economy, as the country is the world's largest producer of cork. For that reason the development of new applications for that material would have a positive impact to the Portuguese economy.

2. Characterization of the components

In order to characterize the different components of the final composite, several test methods were developed to obtain some mechanical and chemical properties of these components, allowing a comparison between them.

For the development of the experimental procedures were considered as main reference ASTM standards. However, in many cases, the application of the standard is not straightforward. Since, in contrast to metals, composite materials have a wide range of combinations that result in different mechanical behaviours.

2.1. Cork agglomerate

Characterization of cork agglomerates was made by both tensile and compression tests in two different types of agglomerate, NL20 and NL11, both supplied by the manufacturer *Amorim*.

2.1.1 Tensile tests

For tensile tests it was necessary to develop methods to provoke failure in the agglomerate.

In the case of axial and tangential directions, production process of the test specimens is represented by four steps in Figure 2. In this process, aluminium sheets were added to the specimen to provide a structural reinforcement and subsequently bored so as to allow the fitting of specimens in the jig system.

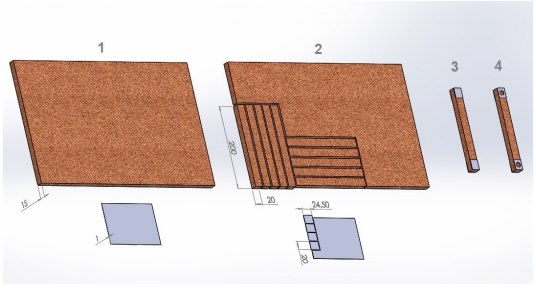


Figure 2: Production process of the test specimens for tensile test on axial and tangential directions

In the case of radial direction, production process of the test specimens is represented by four steps in Figure 3. In this process, T" pieces were added to the specimen to provide a support which allowed them to be grasped by the jig system.

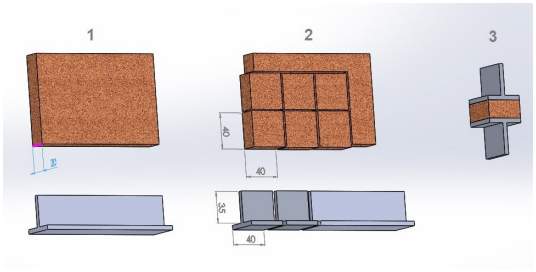


Figure 3: Production process of the test specimens for tensile test on radial direction

After that, specimens were tested at a velocity of 3mm/min. By measuring the applied load and the corresponding apparent displacement, it is possible to calculate the Young modulus of cork agglomerates through the application of Hooke's law. The averages of the Young modulus for each direction tested are summarized in Table 1.

Table 1: Cork agglomerates' Young modulus [MPa]

	Axial	Tangential	Radial
NL20	13,77±0,83	12,67±0,52	9,88±0,03
NL11	3,20±0,47	3,16±0,12	6,02±0,41

For both agglomerates the obtained values are very similar for axial and tangential direction,

which sustains the assumption of a quasi-isotropic behaviour for this material.

2.1.2 Compression tests

As the standard used only refers to the cross-sectional area, for radial direction, 30x30mm NL20 specimens were produced with three different thickness (10, 15 and 20 mm), in order to verify if this parameter has influence on the results. After that, the threes types of specimen were compressed at 5 mm/min.

By dimensioning the test specimens and measuring the applied load and the correspondent apparent displacement for each one of them, it is possible, through the Hooke's law, to calculate de compressive modulus (E_c). The averages of the compressive modulus for each thickness tested are summarized in Table 2.

Table 2: Compressive modulus for the radial direction of NL20 specimens with several thickness

	10mm	15mm	20mm
E_c [MPa]	5,30±0,13	5,34±0,16	5,74±0,31

Observing average values calculated for the compressive modulus, it is not possible to conclude that the specimen's thickness influences this characteristic, as the calculated intervals for the three thickness values tested are superimposed.

After that, both cork agglomerates were tested under the same conditions for two directions: radial and non-radial (representing both the axial and tangential directions). The averages of compressive modulus are summarized in Table 3.

Table 3: Compressive modulus [MPa] for NL11 and NL20 in both directions tested

	Non-radial	Radial
NL20	3,73±0,26	5,34±0,16
NL11	2,51±0,21	2,96 ±0,71

2.1.3 Conclusions - cork agglomerate

It was found that applied methods are good at a comparative level since they allow the characterization of different types of cork agglomerate under the same conditions.

By comparing the values obtained with the values from the agglomerates' data sheets, it was found that for NL20 specimens the results did not present a great discrepancy (11 % for the average tensile strength and 19 % for the average of the compression modulus), whereas for NL11 specimens the results already presented larger discrepancies (42 % for the average strength and 30 % for the average of the compression modulus).

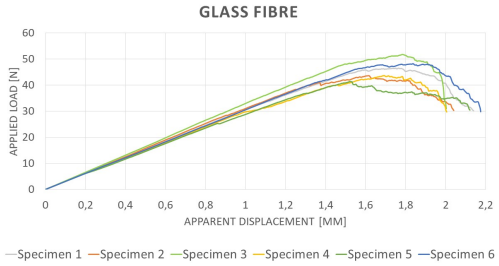
2.2. Glass and carbon fibres

Characterization of the glass and carbon fibres were carried out on both testing the tensile strength of the fibres as well of its woven fabric.

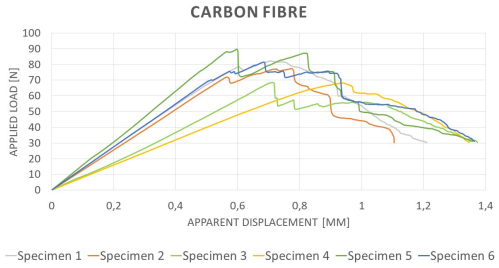
2.2.1 Fibre's tensile test

In order to adapt the test to the jig system available, 50x50mm paper-board reinforcements were added at the ends of the specimens, so that no slipping occurred.

After that, 100 mm long specimens were tested at 3mm/min and, by measuring the applied load and the corresponding apparent displacement, the graphics in Figure 4 were obtained.



(a) Glass fibre



(b) Carbon fibre

Figure 4: Load apparent displacement curves for the fibres' tensile test

Observing the graphics, the difference between glass and carbon fibre rupture is noticeable. The first ones (glass fibres) have an almost immediate rupture, that is, when a first fibre filament fails, the rest also fail almost simultaneously. While in the case of carbon fibres, despite the failure of one of the fibre filaments the remainder continue to offer tensile strength. Due to that, in many cases, after breaking, an increase of the applied load, never reaching the maximum value reached before of the first fault, creating a "steps" effect on the load apparent displacement curves.

By making an approximation of the cross sectional area of each specimen it is possible to cal-

culate the ultimate tensile strength. On the other hand, the calculation of tensile modulus is more complex since it is necessary to make an adjustment due to the rigidity of the testing machine, by calculating the machine compliance. For this, specimens with three different lengths, 100, 200 and 300 mm were produced, so that it was possible to extrapolate the value of C_l, corresponding only to the jig system, that is, when the length of the specimen tends to zero.

The averages of both parameters for each type of fibre tested are summarized in Table 4.

Table 4: Tensile strength [M Pa] and modulus [GPa]

	Tensile strength	Tensile modulus
Glass	854,85±16,68	62,21±10,39
Carbon	982,73±13,71	207,45±25,80

2.2.2 Woven's tensile test

Similar to the previous test, in order to avoid slipping, 50x50mm paper-board reinforcements were added at the end of each specimen, separated by a length of 75mm.

As any cross sectional area approximation that could be performed would have a fairly high associated error, it has been decided, for these specimens, to only analyse the maximum loads, presented in Table 5, obtained by testing the woven at 300mm/min.

Table 5: Maximum load [N]

	Maximum load
Glass fibre's woven	2555,9±72,2
Carbon fibre' woven	4174,0±419,8

2.2.3 Conclusions - fibres

It was found that applied methods are good at a comparative level as they allow the characterization of different types of fibres under the same conditions.

By comparing the values obtained with the ones of fibres' data sheets, it was found that, for both glass and carbon fibre specimens, the properties obtained were relatively close to those presented in the sheets (with discrepancies of 10 and 11 %, respectively) and, in both cases, the data sheet's value is within the estimated range.

With regard to fibre woven fabrics' tensile test, no comparison can be made with the data sheet, as no data relating to the breaking load is presented.

2.3. Epoxy resins

For the characterization of epoxy resins it is essential to fully understand their curing process, as the physical properties of a polymer depend significantly on its degree of cure. Thus, two different resins (CX and AD) were subjected to several tests that allowed to optimize their curing conditions.

2.3.1 Gel time

Gel time is a very useful parameter for manufacturing purposes as it corresponds to the time that a mixed resin takes to become viscous to the point that it is no longer possible to handle it.

The principle of this test is simple. A stamper made from aluminium performs an up-down cycle in a test tube filled with resin. When the point of gelation is reached, the test tube is pulled up by the stamper. This stops the clock which was started at the beginning of the experiment and the gel time can be read.

The test was performed at three temperatures which may correspond to room temperature, i.e. at temperatures at which resin handling may occur. Obtained gel times for the temperatures tested are summarized in Table 6.

Table 6: Gel time at different temperatures

	20°C	25°C	30°C
CX	3h 02min 17s	2h 45min 25s	2h 19min 38s
AD	2h 27min 33s	2h 20min 1s	2h 16min 9s

As it is possible to observe the gel times obtained for the CX resin were, for all temperatures used, higher than those obtained for the AD resin. However, with increasing temperature, this difference decreased due to the high variations of CX resin gel times (approximately between 17 and 26 minutes), when compared to the variations AD resin gel times (between 7 and 4 minutes).

2.3.2 Differential Scanning Calorimetry

The differential scanning calorimetry (DSC) analysis measures heat flow into or from a sample under heating, cooling or isothermal conditions, as a direct function of time or of the sample temperature [6].

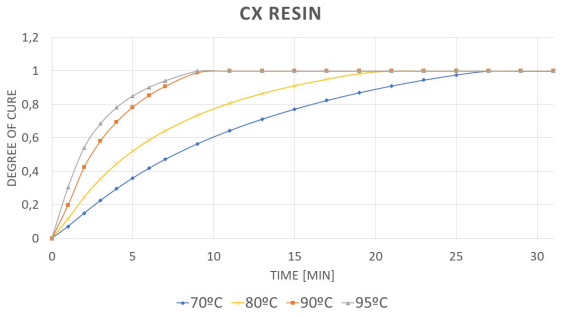
Since the curing process of epoxy resins is an exothermic reaction and the energy involved in the reaction corresponds mostly to the epoxy ring opening energy, it is considered that the exothermic peak observed in the DSC symbolizes the resin curing process. So, this analysis allows to monitor the curing of epoxy resins, and to define the conditions in which it must be carried out.

For the isothermal cure measurements it is possible to estimate the degree of cure as the reaction progresses, using equation 1.

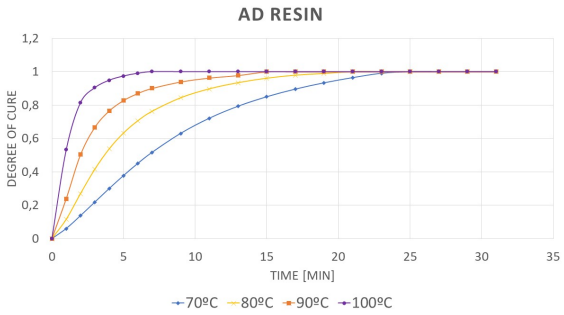
$$\alpha = \frac{H_{rxn} - H_r}{H_{rxn}} \quad (1)$$

Where α is the degree of cure, H_{rxn} equals the total heat of reaction measured at a certain heating rate for an unreacted sample and H_r is the residual heat of reaction for the isothermally cured sample for a certain period of time.

By applying the equation 1 to the heat flow-time curves resulting from the isothermal tests, it is possible to study the evolution of the degree of cure over time at a given temperature (Figure 5 and Table 7).



(a) CX Resin



(b) AD Resin

Figure 5: Evolution of the degree of cure of over time at certain temperatures

Table 7: Time to achieve a degree of cure of about 1, at several temperatures

	70°C	80°C	90°C	95°C	100°C
CX	27	21	11	9	-
AD	25	21	15	-	7

Although the resins appear to be completely cured by the DSC analysis, residual cures and strain relief still have to be optimized during post cure. These processes are more difficult to study through thermal analysis, so they must be studied based on the mechanical properties of the material. For that purpose, it was attempted to produce resin specimens for bending tests, however the results were not satisfactory.

2.3.3 Conclusions - epoxy resins

2.4. Skins

Since it was not possible to study the post cure in resin specimens, this study was carried out on the skins, through tensile tests.

Specimens with dimensions of 200x20 mm were produced in order to contain 60% of fibre volume. After that, they were placed in a press at 70°C and 1 bar during the times established by DSC. After cooling at room temperature, 30x30mm paperboard reinforcements were added at the ends of the specimens. Then, some specimens were placed in the oven at 70C so that the post-cure could occur, while others were directly tested.

The obtained averages and standard deviations are summarized in Figure 6.

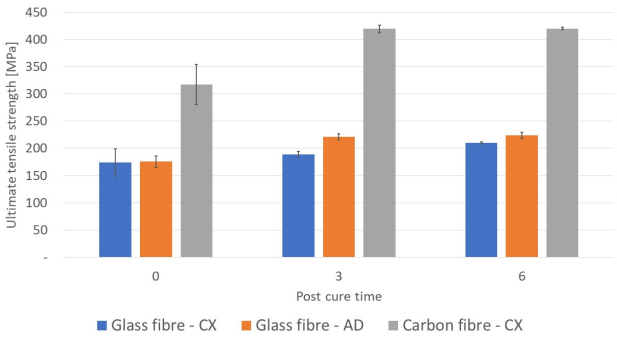


Figure 6: Post cure time effect on the ultimate tensile strength

As can be observed in Figure 6, there was a tendency to increase the average ultimate tensile strength supported by the skins with the application of a longer post cure time. In all cases, the most significant difference occurred between the specimens that were not submitted to any post cure and those that were placed at 70°C for 3h. The difference between the latter and those placed at the same temperature for 6 hours is no longer so relevant, mainly for the specimens with AD resin.

Also noticeable, is a decrease of the standard deviations with the increase of post cure time, showing that the application of this process is important, not only to optimize the properties of the material, but also to homogenize them, which is of extreme importance for quality industrial purposes.

Similar results were obtained from the analysis of the specimens stiffness (Figure 7). This calculation is more complex since it is necessary to make an adjustment due to the rigidity of testing machine, similar to the fibre's tensile test.

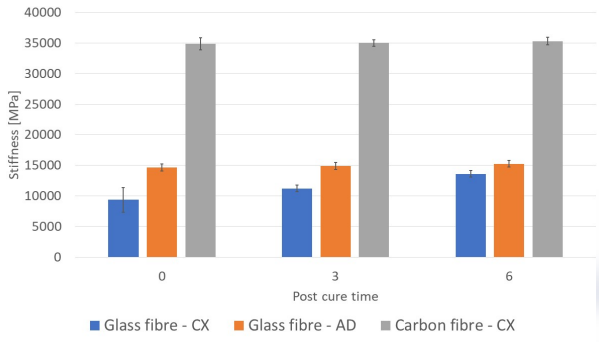


Figure 7: Post cure time effect on the tensile stiffness

2.4.1 Conclusions - skins

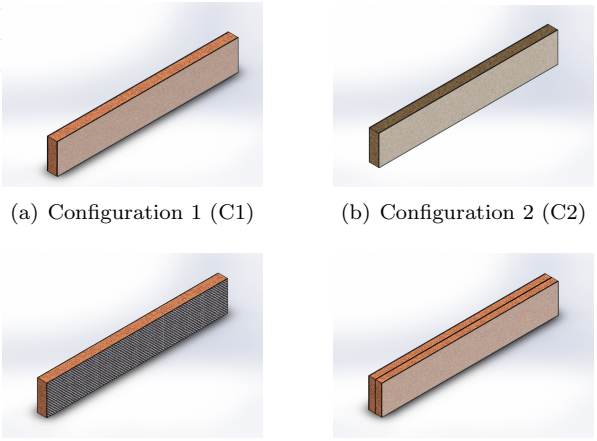
It is concluded that although the applied method demonstrated the effect of post cure on the skins properties, it may not be the most appropriate procedure to study this parameter. This is due to the fact that, in order to evaluate the post cure in this method, a discrete analysis is used instead of a continuous analysis, i.e. curing times are defined previously rather than data being acquired over time.

After these tests, it was decided to continue this study with CX resin. Although the properties obtained for the skins produced with the AD resin were slightly higher than those obtained with the CX resin (on average about 7 and 12% higher), this decision was based on logistic decisions related to a production process in course.

3. Characterization of the sandwich composite structure

In order to understand the influence of each component on the properties of the final composite several fibre-cork agglomerate combinations were developed. The configurations produced by hand lay-up were the following:

- **NL20 - glass fibre (C1)**: conventional configuration with two glass fibre and CX resin skins, separated by a NL20 cork agglomerate core of 15 mm thickness (8 (a)).
- **NL11 - glass fibre (C2)**: conventional configuration with two glass fibre and CX resin skins, separated by a NL11 cork agglomerate core of 15 mm thickness (Figure 8 (b)).
- **NL20 - carbon fibre (C3)**: conventional configuration with two carbon fibre and CX resin skins, separated by a NL20 cork agglomerate core of 15 mm thickness (Figure 8 (c)).
- **NL20 - glass fibre - double (C4)**: configuration that aims to increase stiffness by using a duplicated structure, formed by three glass fibre and CX resin skins, separated from each other by two NL20 cork agglomerate cores of 10 mm thickness (Figure 8 (d)).



(a) Configuration 1 (C1)

(b) Configuration 2 (C2)

(c) Configuration 3 (C3)

(d) Configuration 4 (C4)

Figure 8: Sandwich configurations

After producing the specimens several important characteristics, such as, area density, bending stiffness and production cost, were analysed.

3.1. Area density

In order to evaluate this factor, the various specimens of each of the configurations were weighed and measured, so that an estimation of the respective area densities could be made (Table 8).

Table 8: Area density [Kg/m^2]

C1	C2	C3	C4
46,8 ±0,3	37,4±0,9	47,6 ±0,9	56,4±0,7

3.2. Bending stiffness

After applying the ASTM standard for determination of the flexural properties of a sandwich structures [4] and verifying that the obtained results had no physical meaning, the evaluation of this parameter for each configuration was made through a method based on a linear regression from stiffness as a function of the support span, since this procedure had already been used in cork agglomerate core composites [5].

This method consists of performing several four-point tests with the same loading configuration (Figure 9), but increasing the length of the support span (S=100mm, S=150mm, S=200mm and S=250mm).

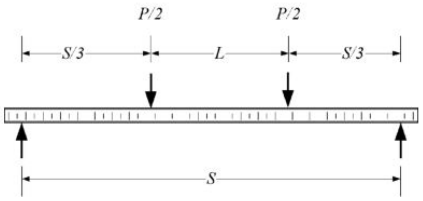


Figure 9: Loading configuration [4]

To ensure that there are permanent deformations in the specimens during tests, the test specimen used for the different support spans must be the same.

Deducing the mid-span equation and replacing L=S/3 (since that corresponds to the chosen loading configuration), the mid-span deflection (Δ) as a function of the applied load (P), bending (D) and shear (U) stiffness can be obtained, which then can be transformed into the following equation (equation 2), similar to the equation of the type $y = mx + q$.

$$\frac{\Delta}{SP} = \frac{1,7}{96D}S^2 + \frac{1}{6U} \tag{2}$$

Plotting the previous equation as a function of S^2, it is possible to determine a linear regression from the obtained results for each sandwich composite configuration, acquiring the mean results for D from the graph slope (equation 3).

$$D = \frac{1,7}{96m} \tag{3}$$

The mean values obtained for bending stiffness (D) for the different configurations of the composite are summarized in Table 9.

Table 9: Bending stiffness [MPa]

C1	C2	C3	C4
17,9±1,3	6,5±0,3	50,4±6,7	28,5±2,9

3.3. Production cost

Production cost is also an important aspect when deciding which configuration to choose.

In order to be able to consider this factor, it is necessary to know the raw materials costs. After that, considering the amount of materials required for each configuration, it is possible to estimate the cost per m^2 of the four configurations (Table 10).

Table 10: Cost [€/m^2]

C1	C2	C3	C4
31,12	33,90	103,85	48,08

In this analysis only raw material costs were considered and production costs (labour, energy, etc.) were not taken into account. However, those represent fixed costs, which will add equal value to all configurations and, for that reason, have no influence for comparative purposes.

3.4. Conclusions - sandwich configurations

Finally, considering the factors that influence the selection of the material analysed previously and using the C1 configuration as a reference, the increase or decrease of the different factors was evaluated in comparison with the values of the same for the configuration C1.

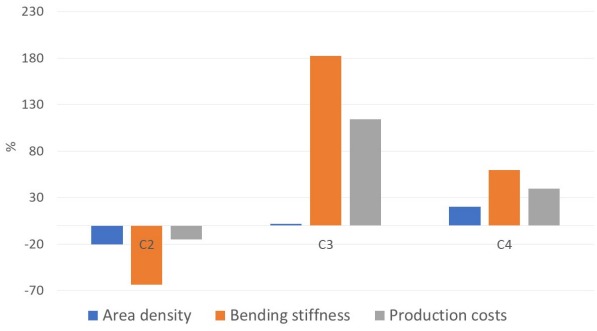


Figure 10: Comparing different configurations with the C1 configuration

In this way it is possible to conclude that the configuration C2 leads to a decrease of the area density, which is an advantage. But at the same time, it also leads to a decrease in almost 70% of the bending stiffness and an increase of the production cost. It is also important to note that in this configuration failure occurred by delamination which is a disadvantage.

With respect to C3 configuration, it has an area density very similar to one of configuration C1, and presents an increase of about 180% of the bending stiffness, which is an advantage for this configuration. However, it also shows an increase of about 230% of the production cost, so it will only make sense to use this configuration in situations where cost is not one of the most important decision factors.

Finally, alternative configuration C4 exhibits an increase in bending stiffness of about 60%, alongside an increase in area density and production cost. Although for this configuration the increase in bending stiffness is not as high as for C3 configuration, also the increase in cost of production has a lower value (approximately 75% less). So this configuration may be an alternative to the carbon fibre resource.

However, it will only be possible to decide which is the best configuration by assigning relative weights to each of the parameters, this assignment

will depend on the intended application.

4. Conclusions

From the studies made through this work it was possible to develop methods to characterize different cork agglomerates, reinforcing fibres and epoxy resins.

Beginning with the cork agglomerates, it was possible to establish methods for both tensile and compression tests. That allowed the analysis of tensile strengths and stiffness and collapse stresses and compressive modulus on both cork agglomerates.

In relation to fibres, it was possible to carry out tensile tests on both the fibres and its woven fabrics. The fibres' tensile test proved to be more accurate, since the values obtained for the tensile stiffness approximated significantly the values of the data sheets of both carbon and glass fibres.

Finally, with respect to the epoxy resins, a method of studying the curing conditions was defined through DSC. However, the study of post-curing parameters has to be performed on the skins through tensile tests.

After the characterization of each component, a method for determining the bending stiffness of the sandwich composite was also established. This allowed to evaluate the influence of the different components on the final composite bending stiffness by using several combinations of fibre-cork agglomerates, which resulted in four configurations. This allowed the development of an alternative configuration (C4 configuration) that, despite not having achieved mechanical properties as high as the configuration with carbon fibre, showed an improvement thereof and a competitive advantage in cost of production.

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