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In this study, green high-density polyethylene (GHDPE) composites with 5, 10 and 15% natural powdered cork (CP) with and without 5% maleic anhydride (PE-g-MA) were developed in order to evaluate the potential of these materials for civil construction applications. The composites were produced in a co-rotating twin-screw extruder and injection molded. An impedance tube was used to determine the acoustic behavior of the composites, which were then used as ceiling tiles in prototypes of provisional constructions in order to study ascertain their performance with respect to thermal comfort in the City of Teresina, PI - Brazil. Our data indicates that composites sound absorption coefficient values are good between 500 and 1200 Hz, especially for those with a higher percentage of cork, with the lowest absorption rate observed for composites containing PE-g-MA. The composites used in the prototypes provided a decrease in the internal temperature of the built structure.

Keywords: Green polymer, sound absorption, thermal comfort.

1. Introduction

Currently, the construction industry is the second largest consumer of plastics in the world, using 19% of world production, losing only to the packaging sector with 42%¹, given the lightness and durability of these materials².

Thermal and acoustic performance are relevant characteristics in polymeric composites used in civil construction and the number of researches that explore such application is increasing. According to the International Energy Agency, energy consumption trends in the world indicate that between the years 1984-2004 primary energy increased by 49% and carbon dioxide emissions by 43%, with an average annual increase of 2.0% and 1.8%, respectively³. The greenhouse effect, caused by increased carbon dioxide emissions, leads to an increase in ambient temperature, which reflects in the internal, external, and surroundings of buildings, causing increased expenses with electricity and fuel. This motivates the search for thermally efficient constructions and stimulates research in the area of polymeric composites⁴.

Acoustic performance is another parameter studied, as noise pollution is a very impacting environmental problem responsible for several types of disturbances, reduced work efficiency and quality of life⁵. The use of materials with good sound absorption in buildings is one of the strategies to reduce noise and increase comfort⁶.

An alternative for such limitations would be to add a natural reinforcement to the polymer matrix in order to obtain a sustainable composite with improved properties^{7,8}. In these composites, both the fillers and the matrices can be from renewable raw materials, biodegradable or recycled sources^{9,10}.

Most green composites are based on lignocellulosic materials, due to the enormous variety of biomass available, which corresponds to the bark of trees, bundles of fibers, leaves or hard fibers, seeds, fruits and cereal straw, among others¹¹⁻¹⁵. Several technical aspects promote the interest in these materials as a supplement or substitution of traditional reinforcements in polymer composites. These include: a) thermal stability to processing temperatures up to 200°C, b) low cost, c) generate of low toxicity waste after incineration, d) are not abrasive to process equipment and e) have specific tensile strength (tensile strength per weight or density) for non-structural applications (partitions, coatings, ceiling, etc.)^{10,16-20}.

Nowadays, there has been a rapid growth in the building sector due to urbanization and a consequence of this expansion

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is an increase in energy consumption and the emission of greenhouse gases, which increases the need for more energy efficient buildings. Conditions of energy efficiency and environmental preservation are dependent on projects and the selection of materials used and the use of composite materials with vegetable loads with characteristics of thermal insulation have been studied extensively in recent years²¹.

Cork is an alternative material for use in polymer composites as it has a similar appearance to wood and in construction industry can be applied in composites with thermal, acoustic and vibration insulation functions (walls, ceilings and floors), false ceilings, cladding, baseboards, mortars, insulating joints and expansion or compression joints, among others^{22,23}. The raw material, besides being a renewable and biodegradable source, is relevant for incorporation in composites²⁴ due to its low density, low permeability to liquids and gases, good compressibility and elasticity, low thermal conductivity coefficient, resistance to mechanical wear, corrosion and fire^{24,26}. Studies show that its main constituents, suberin and lignin, are responsible for many of these characteristics and that these may present antioxidant and thermal stability functions, respectively²⁷.

Among the polymer matrices, polyethylene (PE) stands out for its good processability, high consumption and low cost in the market^{7,28}. This matrix, conventionally of petrochemical origin, can also be produced from the processing of sugarcane, presenting the same characteristics of performance and processability of the fossil origin polymer and can be called green polyethylene²⁹.

The major challenge in the production of polymer-cork composites is to promote good interfacial adhesion between the components²². Cork, which has a polar character, has a low compatibility with nonpolar polymer matrices, especially with matrices made of hydrocarbons such as polyethylene. Thus, the ability of the matrix to transfer stress to the filler through the interface when subjected to mechanical stress is typically reduced^{15,16}.

The incorporation of coupling agents such as functionalized polymers containing maleic anhydride groups in the composition is an effective strategy to improve interfacial adhesion between cork and nonpolar polymers^{22,30}. Other possibilities for increasing the compatibility between filler and polymer are silanization, plasma treatment, hot water and hydroxide based solutions for insertion of chemical groups or removal of contaminants or components of the filler which interfere with adhesion to the matrix³¹. The wide application of these compatibilizers is due to its easy and economic production and the excellent balance of properties achieved while improving the interface between polar and non-polar species, that is, the diversity of connections that can exist simultaneously throughout the interface and in different degrees. Polyethylene functionalized with maleic anhydride (PE-g-MA) demonstrates superior performance compared to other potential polyolefin coupling agents³², in polyethylene composites as its main chain is similar to that of the polymer matrix. Besides, the possibility of a reaction between maleic anhydride and cork hydroxyl groups, favors compatibility.

Thus, the aim of this work was to develop eco-composites of green high-density polyethylene (GHDPE) containing different amounts cork powder (CP) with and without the addition of a polar compatibilizer functionalized with maleic anhydride (PE-g-MA), and to evaluate their acoustic and thermal performance aiming at their use in civil construction.

2. Materials

The polymer matrix used was the green high-density polyethylene (GHDPE) supplied by Braskem, Brazil, *grade* SHA7260, density 0.955g/cm³ and flow rate 20g/10min (temperature 190°C and mass 2.16kg). The vegetal filler used was micronized powder cork (CP) (# 74µm), supplied by Corticeira Paulista Company, Brazil. The coupling agent used, a HDPE functionalized with maleic anhydride (PE-g-MA) Orevac[®] 18507, purchased from Arkema Innovative Chemistry, density 0.954g/cm³, flow rate 5g/10min (temperature 190°C and mass 2.16kg) and melting temperature 128°C.

2.1. Preparation of composites

The powdered cork (CP) was previously oven dried at 80°C for 24 hours, and then mixed with green high-density polyethylene (GHDPE) with and without the addition of a polar compatibilizer (PE-g-MA) through the process of tumbling in proportions indicated in Table 1.

The compositions were processed in a modular twin screw co-rotating extruder, NZ, SJ-20, with a diameter of 22 mm, L/D = 38 and a shape factor of 1.48. The screw used is composed of two sections of intensive mixing formed by kneading blocks. The extruder was operated at 250 rpm and a temperature profile of 160, 170, 170, 180, 180, 180 and 230°C, in its six zones and the head, respectively.

Acoustic absorption samples as per ASTM 1050 Standard with 45 mm in diameter and 18mm thick, were compression molded in a hydraulic press, model MH-08-MN, from MH Equipamentos Ltda., operating under 1.5 ton force, 180°C for 8 minutes.

2.2. Acoustic absorption test

Acoustic absorption coefficients of the matrix and composites were determined in an impedance tube using the two microphone method based on the ASTM 1050 Standard.

For each composition, the values reported are an average of measurements made in three different samples at two different microphone configurations and cutoff frequencies per sample as suggested by ASTM 1050 Standards. See Figure 1 and Table 2.

2.3. Application proposal

Considering the advantages of the materials proposed for the composites manufactured here, as well as the intention of their use in civil construction, the purpose of this work was

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Sample	GHDPE (% by weight)	CP (% by weight)	PE-g-MA (% by weight)
GHDPE	100	-	-
GHDPE/5CP	95	5	-
GHDPE/10CP	90	10	-
GHDPE/15CP	85	15	-
GHDPE/5CP/5PE-g-MA	90	5	5
GHDPE/10CP/5PE-g-MA	85	10	5

Table	2.	Microphone	configurations.
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Microphone 1 configuration	Distance between microphone (mm)	Distance between microphone 2 and the sample (mm)	Low cutoff frequency (Hz)	High cutoff frequency (Hz)
14	120	200	28	1140
24	80	200	42	1715



Figure 1. Microphone position in the impedance tube.

to develop modular plates to be used as a replacement for commercial PVC-based liners in ceilings of temporary sheds in construction sites. Shed prototypes were manufactured as shown in Figure 2.

Prototypes having four different ceiling compositions were assembled with to evaluate ceiling thermal comfort: PVC, GHDPE, GHDPE/5CP and GHDPE /15CP. A commercial flat PVC sheet 8mm thick was purchase and cut to dimensions of 30x20cm. The matrix (GHDPE) and composites plates (15x20cm) with 5 and 15% w/w cork were molded from pellets by means of a MH-08-MN model hydraulic thermo-press manufactured by MH Equipamentos Ltda. Table 3 shows the molding conditions of the plates.

The prototypes containing a sensor for measuring and transmitting internal temperature data to a Klima Logger thermo-hygrometer (TFA-Germany), were placed on a concrete base, in the city of Teresina, at Minister Petrônio Portella Campus of the Federal University of Piauí (UFPI), for 72 hours (06/28/2018 to 06/30/2018). Figure 3 shows the assembly of the prototypes.

Weather Station and thermo-hygrometer temperature measurements were performed every 60 and 15 minutes, respectively. The external and internal temperatures in the prototypes fitted with each type of plate linings was determined, thus allowing the determination of the thermal performance of the prototypes with the commercial GHDPE and PVC.

2.4. Optical microscopy

Optical microscopy was used to analyze the internal structure of the composites from the fracture surface after the tensile strength test. Images were obtained at 40x magnification on a Leica Microsystems MD500 equipment, with ICC 50E capture camera.

2.5. Scanning electron microscopy

The SEM morphological analysis was carried out on the cork particles by metallizing them with a thin layer of gold.



Figure 2. Shed prototype for thermal comfort analysis.

A Shimadzu model SSX-550 microscope with an electron beam accelerating voltage of 15 kV was used for characterization at 50 and 100x magnifications. *2.6. Statistical analysis*

The statistical technique of Analysis of Variance (ANOVA) was used to determine if there is a significant difference between the results presented for each composition. The Tukey multiple comparison test between each pair of averages was also calculated. In this study, a significance level of 5% was adopted

3. Results and Discussion

3.1. Composite acoustic absorption coefficient

Figura 4 shows the acoustic absorption coefficient of GHDPE and its composites under frequencies varying 0 a 3150 Hz.

Data analysis shows the acoustic absorption coefficient to be dependent on the amount of filler in the composite. GHDPE showed a higher sound absorption profile at higher frequencies

Table 3. Molding conditions of the ceiling lining plates used in the prototypes.

Sample	Force (Ton)	MoldingTemperature (°C)	Compression time (min)
GHDPE	3,0	180	8,5
GHDPE /5CP	3,0	180	9,0
GHDPE /15CP	3,0	180	16,0



(a)

(b)

Figure 3. Prototype assembly: (a) fitting of thermo-hygrometer sensors; and (b) placement of the ceiling.

(0.379 at 2500 Hz), while all eco-composites had higher acoustic absorption coefficients than the matrix at low and medium frequencies, up to 1000 Hz, with emphasis on the compositions with the highest cork content: GHDPE /10CP (0.370) and GHDPE /15CP (0.360). These values are, respectively, 25 and 22% higher than that obtained for the neat matrix at the same frequency point (800 Hz). We attribute the improvement in the sound absorption capacity of the composite samples to the high porosity of the cork, since most of the incident sound waves are absorbed and transformed into thermal energy, thus reducing the reverberation²⁶.

Koizumi et al.³³ state that acoustic absorption increases with vegetable filler load. According to these authors, an increase in sample density and friction on the surface of the composite leads to increasing sound waves energy loss and sonorous absorption performance. The improvement in sound absorption coefficient at low frequencies can also be justified according to the studies by Mamtaz et al.³⁴ and Swift et al.³⁵.

For Mamtaz et al.34, factors such as grain size, bulk density and layer thickness of plant loads directly influence the acoustic absorption capacity at low frequencies. Swift et al.35 relate the increase in the absorption coefficient at low frequency with the grain size, stating that flow resistivity is directly proportional to the internal surface area of the granular composite material and that the internal surface area is inversely proportional to grain size. They confirmed that a granular material (2mm in size) contributes to the increase in flow resistivity leading to higher sound absorption performance. Thus, considering that in this study the cork particles used had 74µm, one can conclude that the vegetable filler was responsible for the observed increase in composite acoustic absorption performance and that cork particles significantly contributed to sound dispersion within the material, resulting in a greater capacity to absorb sound waves at low frequency³¹



Figure 4. Acoustic absorption coefficient for GHDPE and its composites.

From 1250 Hz onwards, the sound absorption capacity of the eco-composites was lower than the matrix. This reduction in acoustic absorption performance at higher frequencies is can be assigned to the adopted thickness (18 mm) of the samples. According to the general guidelines for absorption phenomena within a porous material, a long dissipative process of viscosity and thermal conduction between air and the absorbent material within the composite improves absorption. Therefore, sound absorption increasing the sample thickness improves sound absorption^[34].

Berardi and Iannace³⁶ reported a review on the acoustic properties of some natural fillers in their raw state, among them cork. According to these authors, acoustic absorption results for samples of agglomerated cork 10 cm in diameter and 3 cm in thickness was insignificant in the low and medium frequencies, the reason being their grain size. a values of up to 0.9 at 1600 Hz were obtained.

The data obtained here also shows that, in the frequency range investigated, acoustic absorption did not improve by compatibilizer incorporation. This behavior can be justified by the fact that PE-g-MA addition can increase polymer/ filler adhesion and thus decrease composite porosity and internal friction, which disfavors acoustic absorption by the modified composites. This result corroborates the study by Karaky et al.37 on sustainable potato starch composites reinforced with beet pulp (1, 20, 30 and 40%), aimed at applications in insulation coatings on walls and floors in buildings. These authors realized that increased amounts of starch, used as a binder, caused a decrease in composite porosity and on their sound absorption capacity.

The Figure 5 shows the morphological structure polymer composites.

It is important to highlight that, despite having a lower sound absorption coefficient than the matrix (GHDPE) at higher frequencies, GHDPE /15CP still had the highest acoustic absorption coefficient among the composites investigated (0.280). The higher the sound absorption coefficient, the better the sound absorption properties of the material. The best sound absorption properties will be exhibited by a composite material with the most porous filler³⁸.

Figure 6 shows the morphological structure of the vegetal filler added to the polymer, highlighting the cellular profile of cork that influences the performance of composites.

It is important to highlight that the sound absorption coefficients determined for the composites under study



(a)

(b)

(c)



(d)

(e)

Figure 5. Optical microscopy of composites containing cork and compatibilizer: (a) GHDPE/5CP; (b) GHDPE/10CP; (c) GHDPE/15CP; (d) GHDPE/5CP/5PE-g-MA; (e) GHDPE/10CP/5PE-g-MA.



Figure 6. Morphological structure of cork: magnification 50x (a) and (b) 100x.

present similar or better performance rates to some materials applied in civil construction, such as vermiculite (0.14 at 125Hz; 0.50 at 2000 Hz), foamed glass (0.11 at 125Hz; 0.52 at 2000 Hz), foamed plastics (0.03 at 125Hz; 0.85 at 2000 Hz), Glass wool (0.06 at 125Hz; 0.72 at 2000 Hz)³⁹.

3.2. Eco-composites applications

The thermal performance of the prototypes was analyzed and the results obtained are illustrated in Figure 7, which shows both the temperature values at different times inside the prototypes and the external temperature measured by the weather station.

Data analysis points out that all the compositions presented a reproducible behavior during the time period investigated. The highest internal temperatures were obtained in the prototype with PVC plate, both at the times of higher and lower external temperatures. The internal temperatures of the prototypes having GHDPE ceiling plate were lower than those obtained with PVC. The lowest internal temperature on the prototypes were obtained for the eco-composite with higher cork content (GHDPE /15CP). That is to say, internal temperature in the prototypes decreased in the following order: PVC> GHDPE > GHDPE /5CP> GHDPE /10CP. In general, a reduction of 3.44% or 1.4 °C of the internal temperature was achieved for the composite with the highest cork content. The results obtained for the eco-composite boards can be associated with the low heat transfer capacity of cork for the following reasons: the solid fraction is low; the gas contained in the cells has a low conductivity; the cells are small, which eliminates gas convection; radiation is reduced through repeated absorption and reflection in the numerous cell walls²⁵. Table 4 shows the maximum temperatures found in each shed prototype.

The data obtained shows that the external temperatures measured at the A312 weather station are always lower than the temperatures measured inside of the shed prototypes and that, inside the prototypes, the GHDPE/15CP eco-composite displayed the lowest temperature. This indicates that the use of eco-composite plates as a ceiling decreases the internal temperature of the shed, but it does not avoid internal heating by thermal radiation.

Results indicated that the increase in internal temperature occurs rapidly in the early morning in a process that lasts approximately 6 hours, between 6:30 am and 12:30 pm, when the solar irradiation is more intense and the radiation absorbed by the ceiling tiles is transmitted to the interior of the shed prototype. Among other characteristics, cork combines lightness, porosity and low thermal conductivity^{40,41}, corroborating the results obtained for the composite containing

 Table 4. Maximum external and internal temperatures obtained in different test days for each prototype.

Dlaga/agiling	Maximum temperature (°C)				
Place/cening	06/28/2018	06/29/2018	06/30/2018		
Station A312	32,5	32,5	32,6		
PVC	40,3	40,9	40,8		
GHDPE	39,7	40,9	41,1		
GHDPE /5CP	39,3	40,1	40,5		
GHDPE /15CP	38,6	39,1	40,1		



Figure 7. Temperature measured at different times in both insided the prototypes and outside by the weather station: (a) June 28th; (b) June 29th; (c) June 30th.

cork, which showed a tendency of greater thermal inertia than the matrix and the PVC board.

Finally, we can state that the developed eco-composites can be applied as a sustainable alternative in ceilings improving thermal comfort, reducing unhealthy conditions for workers and reducing the costs with mechanical refrigeration. Among other advantages of the proposed material are: the material can be reused or recycled, it is easy to assemble and clean, it has good acoustic performance at low frequencies, it does not require painting, and it is an ecological product. Among the disadvantages found can be mentioned the dark color, which requires internal lighting more frequently and the process of acquiring cork in other states, since it is not a product found in the regional market. According by Aly et. al. (2021)⁴², thermal insulation will play a significant role in reducing energy consumption for construction, especially with the use of ecological materials and agro-industrial waste.

4. Conclusions

The main component of this work was the cork, a material from the porous and fibrous group, with characteristics of thermal and sound absorption, and also of natural origin that favors sustainability in construction. Eco-composites of high density green polyethylene (GHDPE), polyethylene functionalized with maleic anhydride (PEgMA) and powdered cork (CP) as reinforcement material, were developed.

Sound absorption coefficient values indicated that ecocomposites are good sound absorbers at low and medium frequencies, mainly at higher the cork content. PEgMA incorporation increased the adhesion between the phases, reducing the sound absorption capacity of the eco-composites. Prototype sheds having eco-composite ceilings had the lowest values of internal temperature when compared to PVC or GHDPE. Lowest internal temperatures were achieved in the prototype having the eco-composite ceiling with the highest cork content (GHDPE /15CP).

In general, GHDPE /15CP Eco composite results were satisfactory and better than those obtained with either PVC or GHDPE, which indicates that the use of this material as a ceiling with improved thermal and acoustic insulation to be used in civil construction is promising.

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