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Zaida Ortega[1]*, Luis Suárez[2], Jake Kelly-Walley[3,4], Mark McCourt[3]

[1] Universidad de Las Palmas de Gran Canaria, Departamento de Ingeniería de Procesos, Edificio de Ingenierías, Campus universitario de Tafira Baja, 35017 Las Palmas de Gran Canaria, Spain
[2] Universidad de Las Palmas de Gran Canaria, Departamento de Ingeniería Mecánica, Edificio de Ingenierías, Campus universitario de Tafira Baja, 35017 Las Palmas de Gran Canaria, Spain
[3] Polymer Processing Research Centre, School of Mechanical and Aerospace Engineering, Queen's University Belfast, Ashby Building, Stranmillis Road, Belfast, BT9 5AH, United Kingdom
[4] Matrix Polymers, Unit 2, Compass Industrial Park, Spindus Road, Speke, Liverpool L24 1YA, United Kingdom
**Corresponding author. E-mail: zaida.ortega@ulpgc.es*

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MECHANICAL PROPERTIES OF ROTOMOLDED PARTS WITH ABACA FIBER: EFFECT OF MANUFACTURING WITH 1, 2 OR 3 LAYERS

The range of materials suitable for rotational molding is not as wide as for other polymer processing technologies. An option to reduce the carbon footprint of such materials is to introduce natural fibers, such as abaca. In this work, different loadings of abaca fibers (5 to 20 % by weight) were molded using one, two and three-layer constructions. A comparison of the mechanical behavior (tensile, flexural, and impact properties) with the fiber content, considering the method of obtaining the composite (1, 2 or 3 layers) was performed. The thermomechanical behavior of the matrix was not affected due to the introduction of the fibers; apart from a reduction in the storage modulus, especially at low temperature, the curves have a similar profile. In general terms, the tensile and flexural strength were not affected by the incorporation of the fibers, that is, the composites exhibit similar behavior to neat polyethylene. Significant improvements in the tensile modulus were obtained for the parts manufactured with 2 layers, with 10 wt.% fiber in the internal one. As expected, the impact strength was reduced for all the composites, although the layer of PE on the inner side that coats the fibers counteracts this reduction to a certain extent. An increase in the heating time was observed for all the composites made in different layers; although the incorporation of fibers slightly modifies the course of the curve, the heating time is only significantly increased for loadings over 10%. The higher energy consumption needed to obtain the part in the different layers would only then be justified by an increase in the composite properties, which is not the case of the parts obtained in this work.

Keywords: **rotomolding, abaca, cycle time, mechanical properties, DMA, composite**

INTRODUCTION

Rotational molding process is a well-known procedure to obtain hollow parts with good superficial quality and thickness distribution. As the process does not involve any pressure during molding, the obtained parts exhibit null or low internal stresses and high mechanical properties. The main disadvantages of this process are the high cycle time, low energy efficiency of the process and low availability of materials. This last issue is of particular interest nowadays owing to the need to reduce the environmental footprint of plastic products. A strategy to increase the sustainability of rotomolded products is the use of bio- or recycled materials. Several authors have proposed using recycled materials from different sources [1-4], while others focus on the use of waste materials as fillers as a way to valorize industrial residues and reduce the amount of virgin polymer used [5-9]; most commonly inorganic materials used in rotational molding are glass and silicates [10]. Natural fibers have also been proposed in the literature as reinforcement or fillers of rotationally molded sample parts, being agave, sisal, banana and abaca the most studied lignocellulosic materials in this process [11], usually claiming better environmental performance of such composites [12]. Some studies have shown that obtaining parts in different layers [13] can improve the mechanical properties of composites, although some other authors have found no differences between 2 and 3 layer constructions [14-16]. However, not many references about the differences in mechanical behavior because of layering can be found in the literature. Hence, this paper compares the mechanical performance of abaca fiber composites obtained using 1, 2 and 3 layers.

MATERIALS

Metallocene polyethylene (mPE3583UV) from Total Petrochemicals was used as the matrix (density: 0.935 g/cm^3, melt flow index: 8.0 g/10 min). Maleated polyethylene (PEMA) from Dupont (Fusabond EMB226D) was employed as the compatibilizer (density 930 kg/m^3, MFI: 1.75 g/10 min), at 10 % weight with respect to PE weight.

Abaca fiber (grade 2, from Ecuador) was kindly supplied by Celesa (Spain), with a tensile strength of 784 ± 124 MPa and an elastic modulus of 9.5 ± 3.6 GPa, as well as an onset degradation temperature of 270ºC, as obtained in previous works [14, 15]. The thermal stability of the fibers allows their use in rotomolding without any prior treatment; NaOH 1N treatment was in any case applied to the fibers, although this resulted in lower processability of the fibers due to the formation of clusters (results not shown). Nevertheless, it is important to consider whether any chemical treatment applied to the fibers would increase the processing costs and reduce the potential environmental benefits of using such material.

METHODS

Preparation of composites

The samples were produced in a 200 x 200 x 200 mm aluminum mold with a vent hole in a biaxial three-arm carousel rotomolding machine from Ferry RotoSpeed, RS1600 Turret Style model. The rotational speed ratio was set at 8:2, and the oven temperature was set at 300ºC. The oven and peak internal air temperature (PIAT) were constantly monitored using a Rotolog system. All the samples were heated up to about 200ºC (PIAT) before cooling with forced air to 70ºC, followed by demolding. The total weight of material in each sample was 800 g, which allowed 4 mm thick parts to be obtained. The different fabricated moldings are summarized in Table 1. Moldings with treated fiber at the same loadings were also produced.

The abaca fibers were ground together with the PE pellets (or the PE+PEMA) in a ZM200 grinding device from Retsch, with a sieve of 0.5 mm. A homogeneous powder material was obtained following this procedure, as shown in Figure 1a. The prepared material shows irregular shapes of the PE and long fibers (Fig. 1b), with a particle size distribution similar to a conventional commercial rotomolding material (Fig. 1c). This distribution was obtained by sieving the prepared materials in an AS 200 Control Siever from Retsch.

TABLE 1. **Rotomolded parts obtained in 1, 2 and 3 layers**

Structure	Material	Ratio of fiber in final part [wt.%]	Short name
1 layer	PE	0	PE
	PE + 10 wt.% PEMA	0	PEMA
	PE + 5 wt.% fiber	5	PE5
	PE + 10 wt.% fiber	10	PE10
	PE + 20 wt.% fiber	20	PE20
	PE + 10 wt.% PEMA + 5 wt.% fiber	5	PEMA 5
	PE + 10 wt.% PEMA + 10 wt.% fiber	10	PEMA 10
	PE + 10 wt.% PEMA + 20 wt.% fiber	20	PEMA 20
2 layers	600 g PE / 200 g PE (5 wt.% fiber)	1.25	600/200(5)
	400 g PE / 400 g PE (5 wt.% fiber)	2.5	400/400(5)
	200 g PE / 600 g PE (5 wt.% fiber)	3.75	200/600(5)
	600 g PE / 200 g PE (10 wt.% fiber)	2.5	600/200(10)
	200 g PE / 600 g PE (10 wt.% fiber)	7.5	200/600(10)
3 layers	400 g PE/200 g PE (5 wt.% fiber)/200 g PE	1.25	400/200(5)/ 200
	400 g PE/200 g PE (10 wt.% fiber)/ 200 g PE	2.5	400/200(10)/ 200

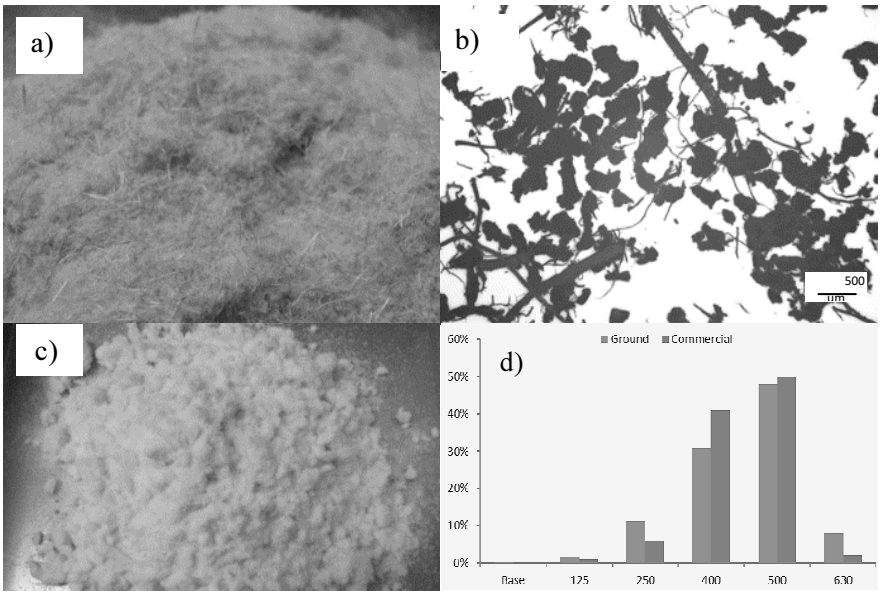

Fig. 1. a) Ground material with 10 wt.% abaca fibers, b) Ground materials and fibers under light microscope, c) Ground material with 10 wt.% treated abaca fibers, showing clusters, d) Particle size distribution obtained by sieving

Characterization of composites

Mechanical testing

Tensile, flexural, and impact tests were performed on specimens cut from the sides of the rotomolded test cubes, according to UNE 527-2:1997, UNE 178:2011, and ISO 6603-2:2000, respectively. An Instron 4411 Universal Tensile Tester was employed for the tensile and flexural tests, with a load cell of 5 kN, at 5 mm/min and 10 mm/min, respectively. At least 5 samples per formulation were used in each test. Impact tests were carried out at room temperature in a CEAST Fractovis instrumented falling weight impact tester, using a total of 10 specimens for each molding, utilizing a dart with loaded total mass of 18.65 kN.

Thermomechanical behavior

The thermomechanical behavior of the rotomolded materials was determined by dynamic mechanical analysis (DMA), performed in a DMA Qseries-Q800, from TA Instruments, from –140ºC to 100ºC, at a heating rate of 3ºC/min, constant frequency of 1 Hz, under the single cantilever flexural mode, with a displacement of 10 μm. Storage modulus (E'), loss modulus (E'') and their relationship (tan δ) were analyzed.

The brittleness (B) of the different materials was calculated as proposed by Brostow et al. [17]:

$$B = \frac{1}{\varepsilon_b \cdot E'} \quad (1)$$

where E' is the storage modulus from DMA at 1.0 Hz and ε_b the elongation at break, both at room temperature.

Differential scanning calorimetry (DSC) assays were performed on the obtained composites, but no significant differences were found either in the melting or solidification temperatures (121 and 112°C, respectively, for the different investigated materials) or enthalpies, and so are not reported in this manuscript owing to extension restrictions. Other authors have found similar behavior for PE-based rotomolded composites, also determining no differences in the crystallinity values of the final product because of fiber incorporation [3, 4]. For these materials, the crystallinity values calculated from the DSC results were found to be between 35 and 40%.

RESULTS AND DISCUSSION

All the rotomolded test specimens with up to 10 wt.% fibers are well consolidated. Besides, they show a smooth external surface and good reproducibility of the mold features (i.e. sharp edges, Figure 1). The internal surface is generally rougher than the external one due to the consolidation of the part in a free environment. In the multilayer structures, the roughness owing to the incorporation of the fibers is responsible for the inhomogeneous flow of the polymer, which leads to the formation of craters. The fibers are well distributed in the surface and the thickness of the parts. The incorporation of 20 wt.% fibers is excessive for the rotational molding process, at least under dry blending as shown in Figure 2a; the volume of fiber used is too much to obtain a good part. Finally, fiber treatment led to the formation of clusters in the internal side of the test parts, but also along the thickness of the parts (Fig. 2b-2d), worsening the aspect of the obtained parts and reducing the processability of the material; only samples with 5 wt.% treated abaca are considered to provide acceptable moldability.

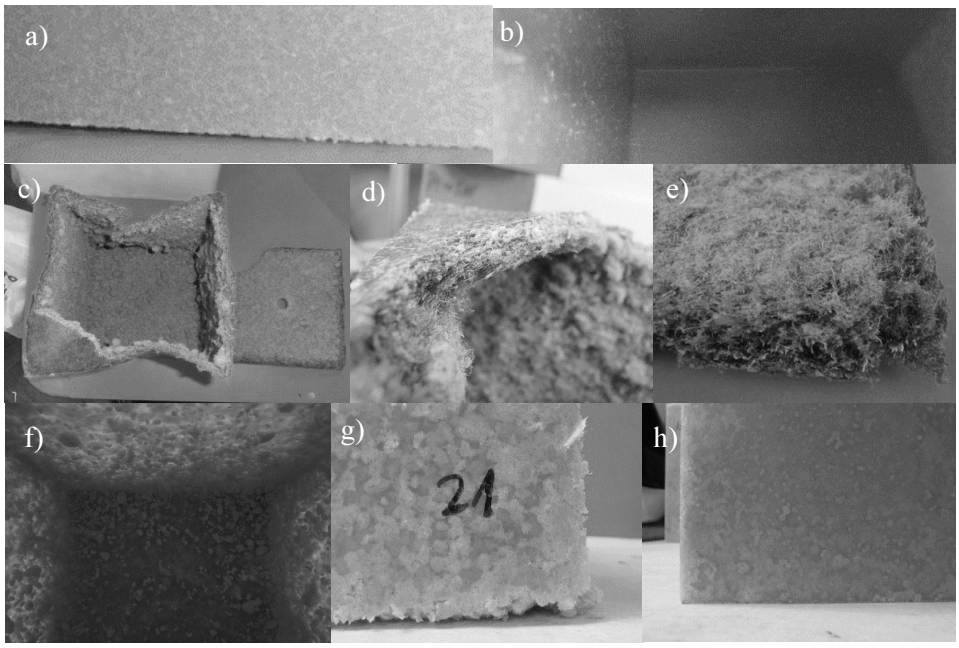

Fig. 2. a) External view of rotomolded cube with 5 wt.% abaca fiber (PE5), b) internal surface of 200/600(5), c) to e) parts obtained with 20 wt.% abaca fibers (failed moldings), f) parts with 10 wt.% treated fibers internal and g) external side, h) external surface of test specimen with 5 wt.% treated abaca fiber (in 1 layer)

Rotomolding cycle times analysis

The oven temperature was set at 300°C for all the moldings, regardless of the number of layers used in each part. Figure 3a shows the readings of a thermocouple placed outside the mold. As observed, the cycle time for one layer is significantly shorter than for two- and three-layer parts for processing the same amount of material (800 g in the final part). This is an issue to consider as longer cycle times are directly related to higher energy consumption, but also to higher possibilities of fiber degradation or polymer oxidation.

The ratio of time dedicated to each stage in the rotomolding cycle is shown in Table 2, considering the total time as the time from the cycle start point to cooling at 70ºC, when the part was demolded. It can be observed that the 5 and 10 wt.% moldings increase the total cycle time by 6% (about 3 min), while the 20 wt.% part increases more significantly, up to 18%. If the different stages are observed, the induction time increases in all the moldings with fibers, as also does the first cooling stage. On the other hand, the sintering stage seems to be reduced, while densification does not reveal any particular trend. The reduction in sintering time can be a consequence of having a lower amount of melting material (polymer). The increase in the induction stage could be attributed to the isolating effect of the fibers, hindering heat transmission. The densification step only shows significant rises for the PEMA10 and PEMA20 formulations; this stage corresponds to the removal of air trapped within the melt and, as observed in the obtained final parts, this was not completely achieved for these parts, which showed a high number of voids and bubbles, even not being able to lead to a consolidated part in the case of PEMA20 (the same happened for PE20, where the fiber was completely degraded). This increase in porosity can be due to the lower MFI of PEMA.

Figure 3b presents the readings of the thermocouple placed inside the mold for the parts made with one layer using PE. It can be observed that the introduction of fibers, even at low loading such as 5 wt.%, provokes a delay in the induction and sintering time, reaching the desired internal temperature (about 200ºC) at longer times with the higher fiber content. This is observed for all the moldings, regardless the use of PEMA as the compatibilizer. Fiber treatment does not have any influence on the total cycle time in the duration of the different stages.

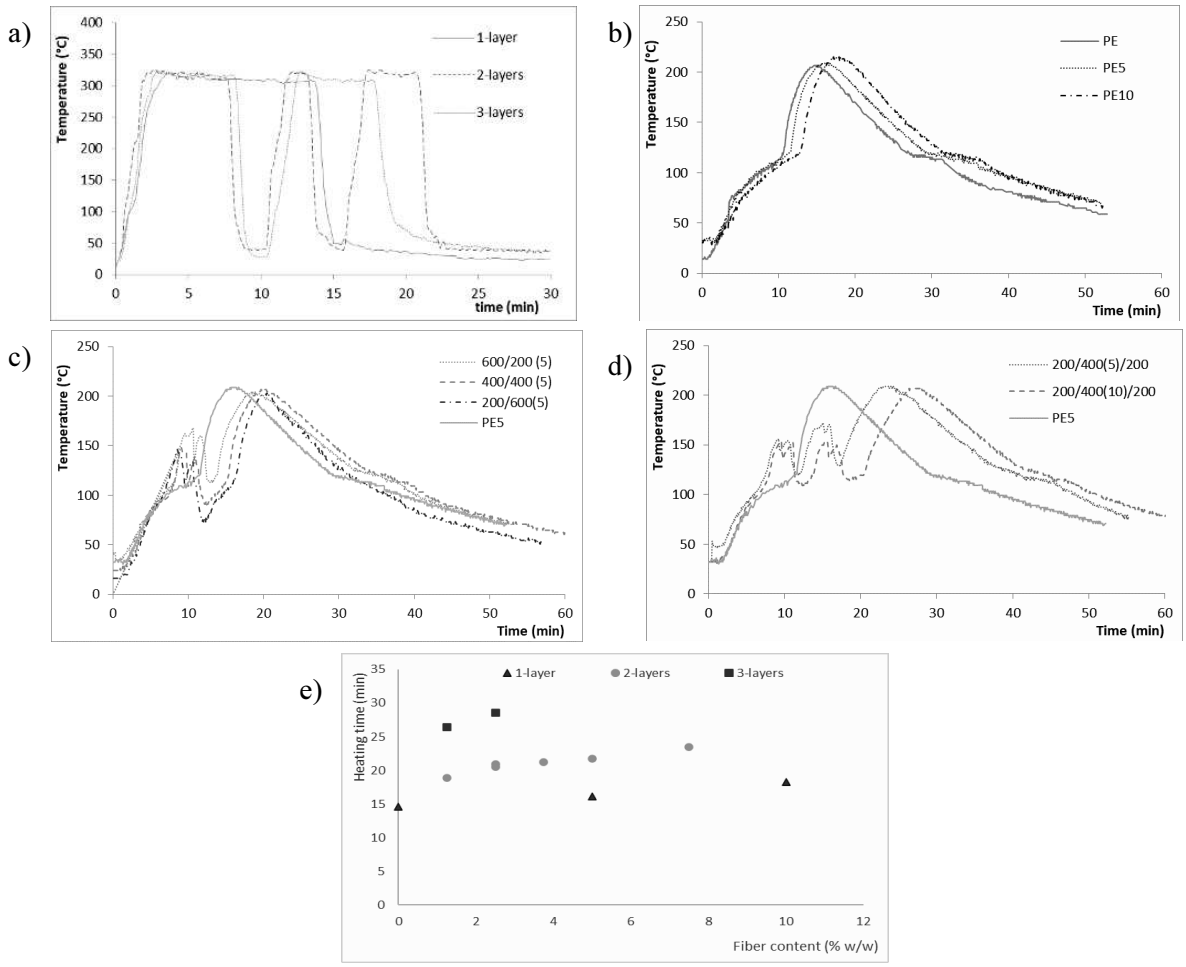

Fig. 3. a) Thermogram for external thermocouple for 1-, 2- and 3-layer parts. Thermograms for internal air in 1 (b), 2 (c) and 3 (d) layers, e) Comparison of heating time vs. fiber content for the 1-, 2- and 3-layer moldings

TABLE 2. **Total cycle time (in minutes) and duration (in %) for processing cycles of specimens produced in single layer**

Stage	PE	PE5	PE10	PEMA	PEMA5	PEMA10	PEMA20
Induction [%]	5.6	8.2	10.4	14.2	15.9	21.4	18.0
Sintering [%]	13.9	12.3	12.5	7.8	6.4	6.6	7.3
Densification [%]	8.9	9.7	9.1	9.0	9.1	12.6	15.1
Cooling [%]	26.1	28.5	26.7	31.0	27.9	27.9	29.2
Crystallization [%]	6.9	8.6	8.7	8.6	8.3	11.6	13.5
Cooling [%]	38.6	32.7	32.6	29.4	32.4	19.9	16.9
Total cycle time [min]	47.2	50.9	51.4	46.8	50.6	50.1	55.2

For the parts obtained in the two-layer structure, the graphs obviously show two peaks of maximum temperature, one for each layer (Fig. 3c). In this case, there are no significant differences in the total time of the three cycles in the different stages (total values). The only differences found in the temperature graph result from the different amount of material used in each layer. As expected, the formulation with the lower amount of material in the first layer reaches the melting point quicker than the one with the higher amount; for the second heating, the trend is repeated: the formulation with the higher mass is slightly delayed with respect to the other ones. The same behavior is obtained for the composites with 5 and 10 wt.% fiber loadings.

When analyzing the curves for the three layers, similar conclusions arise: the higher the fiber content, the longer the cycle times, and the longer the heating time required (Fig. 3d). Finally, the chart of the heating time versus the final fiber content (Fig. 3e) allows the conclusion to be drawn that the effect of working with layers is more significant in terms of energy consumption than the fiber content. Only for the 3-layer parts the total heating time is increased (about 10%); parts with 1 and 2 layers need a similar total cycle time. Hence, taken into consideration the longer heating times, and thus higher energy consumption, together with the difficulties to obtain the moldings in different layers compared to a single one, this strategy would only make sense as a way to increase the amount of foreign material (which is not the case), or as a way to improve the final properties of the part.

Characterization of composites

Mechanical properties

The mechanical properties of the PE/abaca rotomolded composite parts have been reported in previous works of the authors [14-16]; nonetheless, analysis of the influence of the different layers on the mechanical and thermomechanical behavior was not conducted, neither was the relationship between the total fiber loading and the mechanical properties. Figure 4 presents the relationship between the tensile and flexural elastic modulus with the fiber content; the tensile elastic modulus only increases for the parts in the 2-layer structure with fibers at 10 wt.%, remaining unchanged for 1- and 3-layer parts, regardless of the fiber content or the use of PEMA. The fact that only these parts provide an increase in the tensile modulus can be due to the 2-layer structure with 5 wt.% fiber, which does not have enough fiber to achieve this reinforcing effect. Besides, having the fibers concentrated in a layer with an outer skin of well-consolidated homogeneous PE provides better results than having a similar amount distributed in the entire part thickness. In any case, it is interesting to highlight the fact that no decrease in the tensile or flexural strength is seen owing to the incorporation of up to10 wt.% fibers.

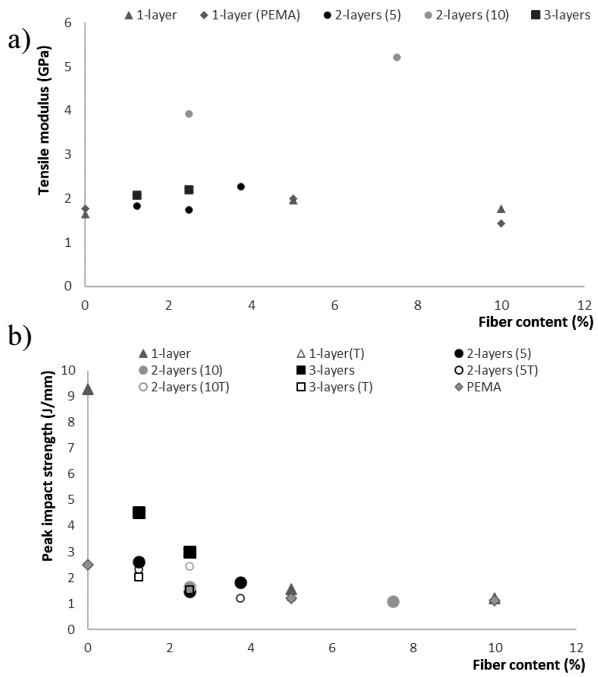

Fig. 4. Variation in: a) tensile elastic modulus, b) peak impact strength with fiber content

Contrary to what was expected, the introduction of PEMA not only does not the increase the mechanical properties, but even reduces them; the 2-layer part with 5 wt.% fiber raises the flexural modulus compared to PE+PEMA, but only reaches a similar value to the one found for PE with 5 wt.% fiber, thus not achieving any positive effect. The tensile and flexural strength exhibit similar behavior to the elastic modulus, that is, there are no significant changes for the composites with different fiber loadings or layers. Only the ones containing PEMA and the composites made of 2 layers with

10 wt.% fiber exhibit lower values of tensile and flexural strength. Particularly surprising was the behavior found for the parts with PEMA; this additive was used as it is well known that it acts as compatibilizing agent between PE and the lignocellulosic fibers. The reduction in the properties can be attributed to the growth in porosity of these samples. Finally, the impact strength is reduced for all the composites, regardless of the amount of introduced fiber; the 3-layer structures seem to provide a lower reduction than the 2-layer ones at the same fiber loading. Therefore, the coating of the fibers with the inner PE layer reduces to a certain extent the negative effects produced by the incorporation of fibers. Fiber treatment does not significantly modify the impact strength of the composites, although the improvement found for the 3-layer structure disappears (Fig. 4b).

Figure 5 displays the cross section of parts obtained by employing the different approaches, with the fibers completely distributed across the section in the 1-layer parts (Fig. 5a), and clear differentiation for the remaining two, with PE in the external side (Fig. 5b) and in the internal and external sides of the composite core (Fig. 5c). In this latter, porosity and a lack of adherence between the layers is clearly visible.

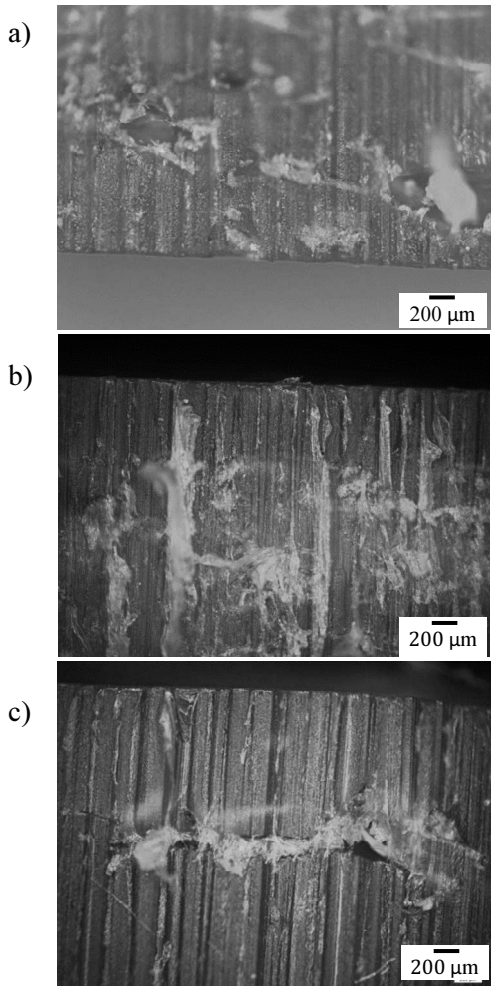

Fig. 5. Micrographs taken on cross section with light microscope for different samples: a) PE5, b) 200/600(5), c) 400/200(10)/200

Figure 6 summarizes the mechanical properties (tensile and flexural properties) for the different parts. It can be observed that some moldings provide the same behavior as the neat PE, while some others can significantly improve the elastic modulus, although with a reduction in tensile strength. The highest tensile modulus was obtained for the 2-layer structures with 10 wt.% untreated fiber or 5 wt.% treated fiber. The worst behavior was obtained for the 2-layer structures with 10 wt.% treated fibers; as already mentioned, the use of treated fibers is limited to 5 wt.%, and higher loadings result in the formation of clusters, which lowers the mechanical performance. The flexural properties are not improved by the incorporation of fibers; as for the tensile tests, the use of treated fibers results in the lowest mechanical properties.

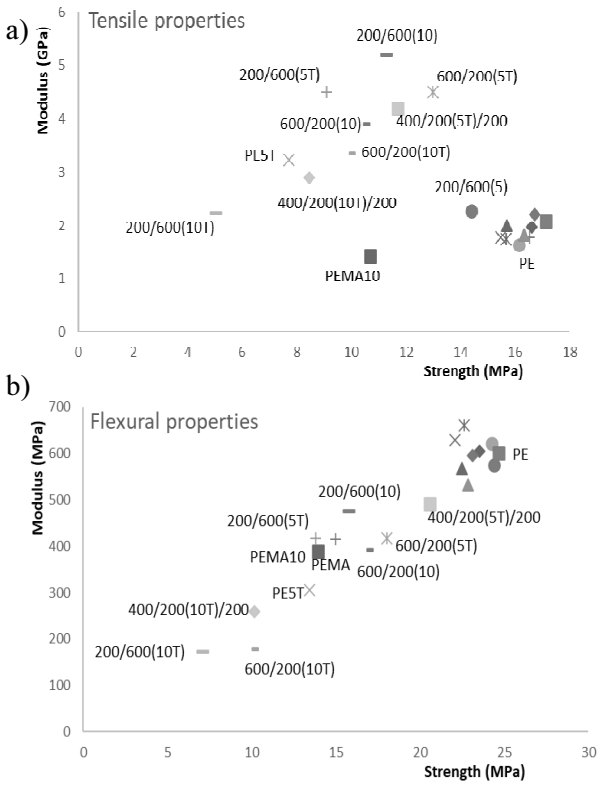

Fig. 6. Tensile (a), flexural (b) properties of different composites

Hence, considering the obtained results, the single-layer structures seem to provide more benefits than using different layers: shorter cycle times, easier molding, higher ratios of used fibers. The use of 2 layers would only be recommended if a high tensile modulus is required (with the inner layer at 10 wt.% loading), while 3 layers (400/200(10)/200) would be applied to reduce the drop in impact strength. Fiber treatment is discouraged, at least for dry blending.

Thermomechanical behavior

Dynamic mechanical analysis (DMA) is an essential technique to assess the mechanical properties of composites because it allows the viscoelastic properties to be studied at different temperatures and it is sensitive to

structural changes as well as interfacial adhesion between the fibers and the matrix [18]. The analysis of storage modulus (E′) as a function of temperature allows the changes to be evaluated in the mechanical properties due to the addition of fibers in different ratios. In this case, all the composites exhibit a lower storage modulus than neat PE, which means that the fibers do not provide any reinforcing effect along the range of the studied temperatures, although the differences are reduced with the increase in the temperature (Fig. 7a). The parts containing PEMA also exhibit a decline in the storage moduli when compared to neat PE. Only for the composites containing PEMA, both at 5 and 10 wt.%, the fiber provides some reinforcing effect, although at temperatures over –10ºC.

The values of storage modulus decrease in an expected constant way, as also found in other works [19, 20]. Other authors working with a blend of HDPE and PEMA have found increases in the storage modulus at different temperatures; for example, the use of 20 wt.% microcellulose fibers increased the storage modulus at –20ºC by about 45% [21], while for 20 wt.% hemp these increases reached 30% and 100% at 0 and 75ºC [22]. In both works, the composites were obtained by injection molding, and so the results cannot be directly comparable. Lei et al. found an increase in both the storage and loss modulus when using bagasse or pine flour in a recycled HDPE matrix, for a 30 wt.% loading [23], although no further improvements were obtained by using PEMA as the compatibilizer, which also happened in this study. The reduced stiffness correlated with the reduction found in the tensile and impact tests and with the higher values obtained for tan δ.

The tan δ plot of PE and PEMA (Fig. 7b) reveals a peak in the curves at about 60ºC, corresponding to α-transition. The decreased stiffness found for the composites implies an increase in the damping properties compared to the neat PE, as seen from the tan δ plot, particularly at low temperatures. Good fiber-matrix adhesion would limit the mobility of polymer chains and thus reduce the damping factor [24]; thus, both the static and dynamic tests suggest that the fibers are not well bonded to the matrix, even with the use of PEMA.

Regarding the use of layers, the curves are closer to the neat PE than for the single layer samples because of the reduced content of fiber (Fig. 7c). Similar results were found for all the composite specimens, despite the number of layers or the fiber distribution in those layers. The 3-layer parts show a less smooth curve, which might be due to the irregular distribution of the internal PE layer. It can be concluded that the thermomechanical behavior of the composites does not show any significant differences, regardless the number of layers or the weight of fiber used, as also observed by other authors [19].

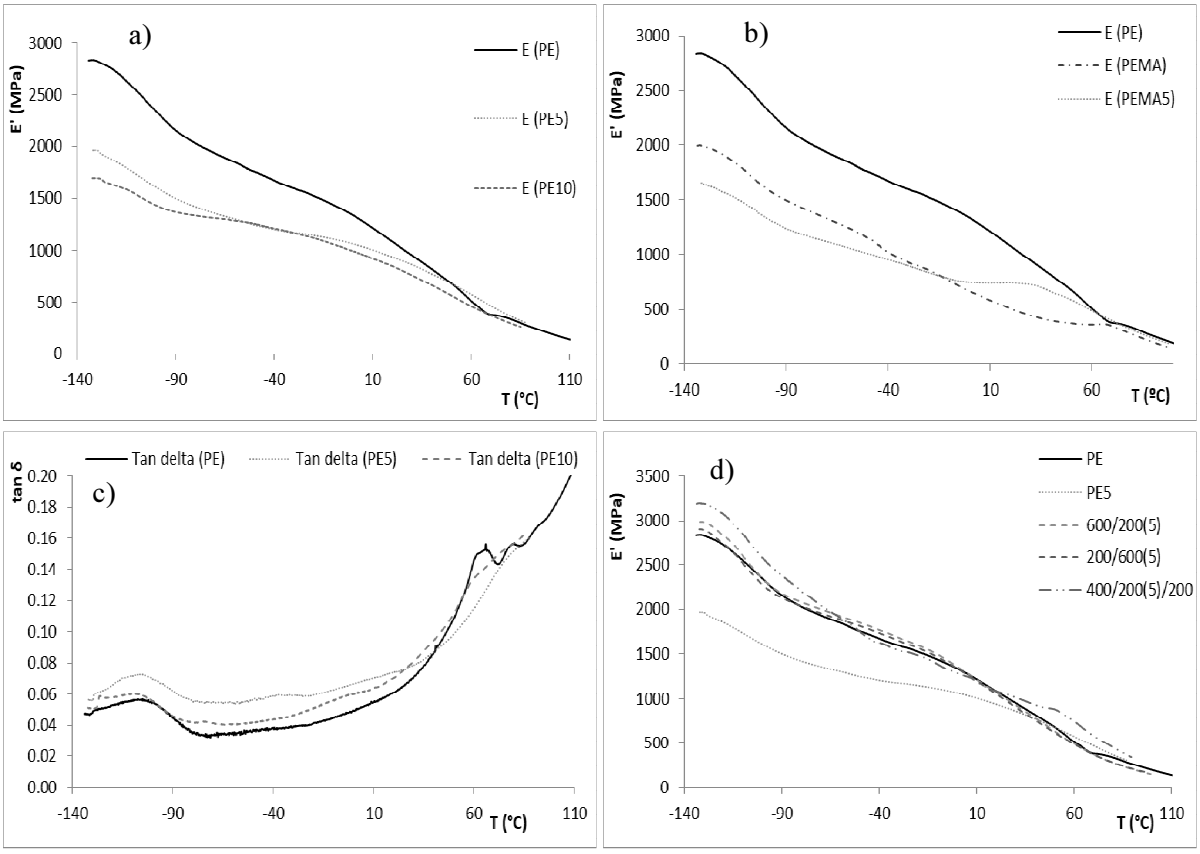

Fig. 7. Storage modulus versus temperature (a, b) and tan δ vs. temperature for specimens (c) made in single layer, d) storage modulus for 5 wt.% composites in 1, 2 and 3 layers

TABLE 3. **Summary of results from DMA testing at 25°C (E' – storage modulus, ε_b – deformation at break, B – brittleness, tan δ – damping factor, A – adhesion factor)**

Material	Fiber content [wt.%]	E' [10^9Pa]	ε_b [%]	B [10^{-9}Pa^{-1}%$^{-1}$]	tan δ	A
PE	0	1017.49	197.64	4.97275E-06	0.065186	0.0000
PEMA	0	477.21	53.48	3.91831E-05	0.079437	0.2186
PE5	5	898.12	21.02	5.29702E-05	0.076184	0.1174
PE10	10	803.83	17.69	7.03249E-05	0.075347	0.0537
PEMA 5	5	734.42	17.60	7.73646E-05	0.084200	0.2350
600/200(5)	1.25	998.05	54.76	1.82972E-05	0.066158	0.0038
400/400(5)	2.5	989.14	24.33	4.15526E-05	0.069744	0.0465
200/600(5)	3.75	978.04	20.97	4.87577E-05	0.071813	0.0654
600/200(10)	2.5	903.00	21.78	5.08455E-05	0.071763	0.0768
200/600(10)	7.5	1079.55	20.51	4.51639E-05	0.070105	0.0044
400/200(5)/200	1.25	1056.92	17.15	5.51688E-05	0.065952	0.0007
400/200(10)/200	2.5	1161.76	21.18	4.06405E-05	0.066163	–0.0072

The brittleness and adhesion values obtained at 25ºC for the different formulations are found in Table 3. According to the literature [17, 25], lower values of brittleness imply higher dimensional stability of the material under repetitive loading; it can be observed that all the composites are one order of magnitude higher than the neat PE (even PE+PEMA), that is, the composites are less stable than pure PE. In any case, a lower brittleness value for the composites was obtained by the composite made of two layers, with the lower fiber content (600/200(5)). The remaining ones have similar values among them, lower for the 2-layer structures than for the ones with 3 layers; these formulations also provided better mechanical behavior in the static mechanical testing. Similar conclusions arise from the adhesion factor, that is, the formulation 600/200(5) shows a lower adhesion factor, which means that higher interfacial adhesion is obtained. The incorporation of higher fiber loadings results in a poor interface, explaining the obtained lower mechanical properties.

CONCLUSIONS

Abaca fibers were successfully rotomolded in up to 10 wt.% loadings without any melt compounding operation, obtaining parts with good aesthetics, fiber distribution and reproduction of parts. Higher loadings did not allow consolidated parts to be obtained.

The impact properties were drastically reduced because of the introduction of materials different to PE as fibers or even PEMA. Furthermore, the use of PEMA as the coupling agent does not improve the properties of the composite.

The use of 3-layer structures counteracts the effect of introducing the fibers in the impact properties, obtaining better behavior than for the specimens with 2 layers for the same loading of fiber.

The tensile elastic modulus only increases for the 2-layer parts with fibers at 10 wt.%, remaining unchanged for 1- and 3-layer parts, regardless of the fiber content.

No significant differences in the thermomechanical behavior were found owing to the fiber incorporation, apart from a reduction in the storage modulus at low temperatures.

The cycle time for one layer is significantly shorter than for 2- and 3-layer parts, although the heating time is only increased (about 10%) for 3-layer structures.

Considering the obtained mechanical behavior, and the increased energy consumption associated with longer cycle times, 3-layer structures are only recommended if the impact strength reduction as a consequence of fiber use needs to be shortened, and 2-layer structures (with 10 wt.% fiber in the inner side) were only used in the case of achieving a high Young's modulus, without reducing the tensile strength and flexural properties. One-layer parts with 5 and 10 wt.% untreated fibers provide similar mechanical behavior to neat PE, and therefore constitute an interesting alternative to produce parts with a wood-like aspect and a potentially lower environmental impact. In any case, a lifecycle assessment should be performed in order to analyze the potential effect of the partial substitution of PE by a lignocellulose filler of a renewable nature.

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