DEVELOPMENTS TOWARDS A MORE SUSTAINABLE ROTATIONAL MOULDING PROCESS

M.D. Monzón[1], Z. Ortega[2]*, A.N. Benítez[2], F. Ortega[1], N. Díaz[1], M.D. Marrero[1]

[1]*Dpto. Ingeniería Mecánica, Universidad de Las Palmas de Gran Canaria, Campus de Tafira, 35017, Las Palmas de Gran Canaria, España*
[2]*Dpto. Ingeniería de Procesos, Universidad de Las Palmas de Gran Canaria, Campus de Tafira, 35017, Las Palmas de Gran Canaria, España*
*zortega@lfi.gi.ulpgc.es

Keywords: banana fiber, rotomoulding, natural fiber.

Abstract
Rotational molding is a method to produce hollow articles with relatively low internal stresses, being an economic alternative to other process such as blow or injection molding. However the availability of materials is limited to few plastics due to small market and difficulties in processing all materials suitable to be applied in other processes. An example of this is the use of natural fibers and biodegradable polymers, well known in injected plastic parts. This paper focuses on applying these new materials in rotational moulding for enabling high quality of natural composites and a more sustainable process. Natural fibers, extracted from the banana plant, are mixed with polyethylene under different ratios. Several mechanical tests and characterization of the processed samples with this natural fiber were carried out.

1 Introduction

Rotational molding process is a well know procedure to get hollow parts, with a good superficial quality, good thickness distribution and high mechanical properties, mainly due to the low or null existence of internal stress, because this process does not involves any pressure during the moulding or it is very low. The main disadvantages of this process are the high cycle time, the low energy efficiency of the process and the low availability of materials suitable to be processed by this technology.

Nowadays, it exists a crescent awareness on the needs to reduce the environmental problems occurred due to industrial processes. This usually undertakes environmental rules and policies, trying to improve that way the global effort to get a more reasonable balance between development and ecology. Among other aspects, the use of new formulations of plastic materials, environmentally more friendly, are being deeply studied; this fact includes both the research on biodegradable and on the use of materials from renewable sources, such as natural fibers. The use of biodegradable polymers is currently limited due to their higher prices, lower mechanical and chemical resistance and difficulty of processing. However, companies producing this type of products are developing this king of products improving their characteristics at lower prices, making them an alternative to traditional plastics.

On the other hand, growing potential of natural fibers has practically no limits, restricted only by the growing period of each plant. In the recent years the interest in using natural fibers as reinforcement in composite materials has increased, as part of the strategy to reduce the amount of composites wastes and their impact in the environment. Some works on injection molding with vegetal fibers, as sisal, flax or hemp have been carried out [1, 2], finding a number of different applications [2]. In 2005, 19 000 tonnes of natural fibers, excluding wood and cotton, were used in automotive composites and around 2 000 tons of natural fiber composites were used in other industries in the EU [2]. Injection of flax or sisal fibers reinforced composites have been widely studied [1 – 3], but just a few references in using this type of fibers for rotational molding have been found. In fact, just some references of using reinforcements or any other type of materials, different of plastics, in this process; this is due to the high sensibility of this process to the introduction of foreign materials, reducing in a significant way the mechanical properties of the obtained part, mainly the impact strength [4]. The use of a composite containing 10 % in weight of flax fiber [5] does not modify the tensile neither the impact strength of samples. Composites prepared with cabuya and sisal do not improve the tensile properties of polyethylene and a significant reduction in impact properties occurs [6]. Furthermore, the use of foreign materials, such as fibers, is that the process is low shear and good mixing of resin and reinforcement is not good at these conditions, happening that fibers segregate out of the powder, laying on the inner part surface [7]. In this context, a novel research work is aroused to study the behavior of composites of natural fibers in rotational molding, as their characteristics [1 – 3, 8 – 10].

This study focuses on the use of fibers obtained from banana and abaca plants. These fibers are considered as hard fibers, and they belong to *Musa genre*, *Musaceae family*. Banana plant's specie is *Musa paradisiaca* while abaca belongs to *Musa textilis* specie. Banana is the first crop in Canary Islands, regarding to the production numbers, and the second one in cultivated area [8]. The use of this fiber, obtained from the waste of banana food production, in composites industry means a revaluation of this crop in the Archipelago. Nowadays around 9 562 ha of banana trees are cultivated in Canary Islands [8], with a potential in fiber production of about 25 000 tons per year. Abaca is grown in Philippines in about 130 000 ha, but also in other Southeast Asia and Ecuador, which is the second producer of this type of fiber. In 2007 Philippines produced 60 000 tons of abaca fiber and Ecuador, 10 000 tons [2].

2 Materials and testing methods

Banana fibers were extracted at Universidad de Las Palmas de Gran Canaria [8] and abaca fiber was obtained from Celesa (abaca from Ecuador). Polyethylene (PE) used was kindly supplied by TOTAL Petrochemicals (Lumicene® M3581UV). Both fibers were characterized by means of different techniques. Obtained parts were tested to find their mechanical properties.

2.1 Mechanical properties of fibers

Several single fibers were tested in an Instron 5564 test machine, at a rate of 1mm/min, in the Polymer Processing Research Centre (PPRC) facilities, at Queen's University of Belfast.

2.2 FTIR studies

FTIR studies of both fibers fiber were made at PPRC, in a Perkin Elmer Spectrum 100 FTIR Spectrometer with attenuated total reflectance (ATR) device, collecting 60 spectra per second

at a resolution of 16 cm^{-1}, with wavelength from 4000 to 500 cm^{-1}, obtaining the spectrum in terms of transmittance versus wavelength.

2.3 Thermal stability

Thermogravimetric analyses (TGA) were run to determine the thermal degradation of fiber. A Mettler Toledo TGA/DSC 1 analyzer was used at a heating rate of 5ºC/min in both nitrogen and air atmosphere (10 ml/min). Isothermal studies were also carried out by putting the fibers at 220 ºC for 60 min, in both atmospheres (10 ml/min of gas flow).

2.4 Optical microscopy

Observations of samples were carried out in an Olympus BX51 optical microscope, at different magnifications and under polarized light.

2.5 Preparation of formulations

Different formulations were prepared, using milled fiber, produced at a ZM200 milling machine. Material containing 5, 10 and 20 % of fiber (abaca or banana) was used for the production of rotomolded parts.

2.6 Rotational molding

Rotational molding tests were carried out at PPRC in a carousel biaxial rotomolding machine from Ferry industries.

2.7 Final parts testing

Tensile, flexural and impact tests, as well as aesthethics, were made to find the properties of the obtained parts. Tests were performed according to ISO standards: 527 for tensile, 178 for flexural and 7765 for impact tests. An Instron 4411 Universal Tensile Tester was used for tensile and flexural tests (at 5 mm/min and 10 mm/min, respectively). Impact tests were performed on a Ceast Fractovis Free Falling Dart machine, with a total weight of the dart of 18.63 kg at room temperature.

3 Results

3.1 Optical microscopy

Banana and abaca fibers were observed at different magnifications, under normal and polarized light. Banana fiber has an average diameter about 180 μm (figure 3), while abaca shows a diameter about 381 μm. Different structures can be observed in the picture, as the light refracts in different ways. These differences in light refraction might be due to single microfibrils existing in the fiber, bonded together by pectin and other non-cellulosic components. Diameters of microfibrils measure 11 to 17 μm, for both types of fibers. Abaca fiber also shows a higher dispersion of values, even for the same measured fiber.

3.1 Mechanical properties of banana fiber

Up to 20 samples of fibers were tested, obtaining their stress – strain curves. Results show a tensile strength of 1032 ± 110 MPa and an elastic modulus of 37 ± 3 GPa for banana fiber and 700 ± 151 MPa and 24 ± 6 GPa for abaca fiber. As can be observed in table number 1, the values obtained for banana fiber are even higher to those obtained for other vegetal fibers. Abaca fiber shows comparable behavior to other natural fibers, and seems to be lower than for banana. This fact can be due to the more uniform section of banana fiber, which allows calculating a more precise cross section, while for abaca fiber a wider distribution of diameters is observed, in the same filament of fiber.

Fiber	Tensile strength (MPa)	Elastic modulus (GPa)	Reference
Banana	1032	36.9	--
Abaca	700	24	--
Flax	802	33	[1]
Banana ties	963	38	[8]
Palm	377	3	[9]
Bamboo	503	36	[9]
Cotton	400	12	[11]
Sisal	575	15	[10]

Table 1. Mechanical parameters for different vegetable fibers

3.2 FTIR analysis

Abaca and banana fiber show very similar IR spectra (figure 1), with the same peaks and transmittance values also similar, confirming that both fibers have a close chemical composition, as expected, as they belongs to the same vegetal genre and family, showing typical absorption bands of vegetal fibers (cellulose, hemicellulose and lignin) [12]. A broad peak at around 3300 cm^{-1} is observed, due to vibration of bonds O – H in organic compounds. The peaks at 2900 cm^{-1} is due to the vibration of C – H bonds in CH and CH_2 in cellulose and hemicellulose. At about 1700 cm^{-1} it is observed another peak, which is mainly attributed to ketones or carbonyl groups in lignin, waxes and pectin. The band at 1430 cm^{-1} indicates the presence of metoxi radical ($O\text{-}CH_3$), associated with hemicellulose in fiber. There are several peaks with weak intensity between 1369 and 1100 cm^{-1}, mainly related to aromatics and C-O in lignin and hemicellulose [12]. The more intense band appears at 1060 cm^{-1} and it is linked to secondary alcohol groups, quite frequent in sidechains of cellulose, hemicellulose and lignin.

The only significant difference between both spectra is in the peak at 1735 cm^{-1} in the abaca spectrum, which is just an inflection in the banana one. This peak corresponds to the vibration of C = O groups (from lignin and waxes), meaning a higher content of these components in abaca than in banana fiber. This can be confirmed by observing the peak at 1260 cm^{-1}, due to C – O bonds in aromatic skeletons, more intense for abaca.

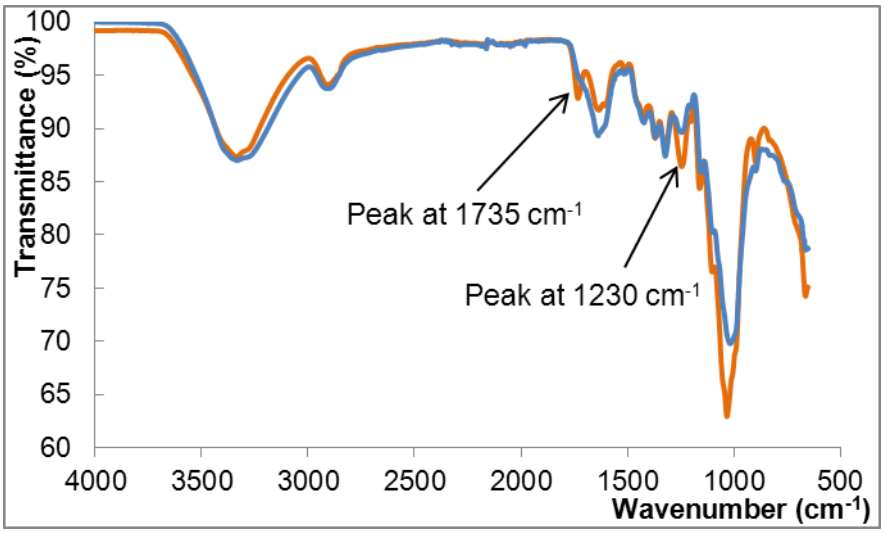

Figure 1. FTIR – ATR spectra for banana (in blue) and abaca (in orange) fibers

Spectrum obtained for banana and abaca fibers are also very similar to those obtained for other authors [1, 9, 11 – 13] for different vegetal fibers, allowing concluding that all these fibers have a very similar chemical composition.

3.3 Thermal stability

There are no significant differences in the thermal behavior of both types of fibers in air neither in nitrogen atmosphere. Table 2 shows the average values obtained in thermal tests. Content in humidity is quite close between both fibers, while residues at the end of the test are more important for abaca than for banana fiber; this could mean that abaca fiber has a higher content in inorganic compounds. The only differences between the tests carried out under nitrogen atmosphere are slightly higher degrading temperatures and the presence of residues; that is totally expected, as organic compounds degrade more in oxygen presence. Isothermal tests also show a very similar behavior for both types of fibers.

		Air		**N_2**	
		Abaca	**Banana**	**Abaca**	**Banana**
Dynamic tests	**Left limit temperature (ºC)**	217	213	243	213
	Onset temperature (ºC)	270	273	303	293
	% Humidity	9.74	8.76	8.94	8.20
	% Ashes	4.21	0.61	13.7	6.50
Isothermal tests	**% Weight loss at the end of the test at 220 ºC**	20	21	14	17

Table 2. Results obtained from thermal tests (dynamic and isothermal)

TGA analysis shows that thermal degradation of banana tree fibers in N_2 atmosphere is about 221 ºC, as that is the temperature were weight loss start, after the moisture in the fiber is removed, (named in table as left limit temperature). Results obtained by other authors show a degradation temperature of 205 ºC for jute, 250 ºC for hemp and 220 ºC for okra [13].

Regarding to residues of fibers at 725 ºC, in air atmosphere flax shows a residue of 1.6 %, while α – cellulose residue is 0 % and lignin is 7.7 % [14]. Banana fiber has a residue at this temperature of 2.8 % and abaca fiber of 1.2 %. This result indicates the similar chemical composition of these fibers, as results obtained are again very close.

Isothermal studies carried out for textile flax show a weight loss of around 4 % in air atmosphere at 230 ºC for 10 minutes [14]. Banana fiber show a similar value while abaca fiber weight lost is about 1 %.

3.5 Preparation of formulations and rotational molding

Different formulations containing abaca or banana fiber were prepared, and are summarized in table 3. All moldings were prepared with 800 g of the material; when molding with fiber, the material was prepared by grinding together the PE pellets and the fiber, avoiding that way the compounding step, which can thermally degrade the fiber, and reducing the thermal cycles to the fiber at just one.

All samples were processed in the same way, with an oven temperature of 350 ºC, stopping the heating time when internal air reaches 210 ºC and cooling until 70 º C. Slight differences

have been observed in the internal air curves obtained, being the most important placed in the cooling stages, concluding that the higher content of fiber in the part the longer cooling time is needed to reach the demolding temperature. Formulation containing 20 % of fiber could not be tested, because it was not possible to get it out of the mold without breaking it.

	PE (g)	**PEMA (g)**	**Abaca (g)**	**Banana (g)**
PE	800			
PEMA	720	80		
PE 5% AF	760		40	
PE 10 % AF	720		80	
PE 5% BF	760			40
PE 10 % BF	720			80
PEMA 5 % AF	680	80	40	
PEMA 10 % AF	640	80	80	
PEMA 20 % AF	560	80	160	

Table 3. Formulations used in rotomolding production of test samples (cubes)

It was also observed that in the internal part of this cube there were a lot of bundles of fiber that did not were incorporated in the matrix, meaning that this amount of fiber is too high. The rest of tests have shown good aesthetics on the external face, with a darker color for the formulations with higher amounts of fiber. Figure 2 shows some of the tests samples produced.

Figure 2. Some of test samples produced (left). PE 5 % BF before molding (upper right) and detail in one of the cubes produced (down left)

Tensile, flexural and impact tests were carried out to all produced samples (except to PEMA 20 % AF). Results (summarized in table 4) show that tensile strength is not improved by the use of fiber; the same happens for flexural strength. However, elastic modulus in tensile tests increases very significantly with the use of fiber, especially for banana fiber; this means that stiffness of the part is improved by the use of banana fiber, getting an elastic modulus up to twice higher than for virgin PE. Impact strength decreases a lot, as expected, because of the inclusion of fibers, even just by adding PEMA. Results in impact tests do not show any significant difference due to the type of fiber or the use or not of PEMA. Best flexural results are obtained for composites with abaca fiber or with just PE.

	Tensile tests		**Flexural tests**		**Impact tests**
	Strength (MPa)	**Elastic modulus (GPa)**	**Strength (MPa)**	**Elastic modulus (MPa)**	**Peak impact strength (J/mm)**
PE	16,14	1,64	24,26	621,86	10,22
PEMA	16,52	1,78	14,99	415,27	2,50
PE 5% AF	16,61	1,96	23,51	605,72	1,55
PE 10% AF	15,48	1,77	22,06	628,20	1,20
PEMA 5% AF	15,69	2,00	22,48	568,42	1,21
PEMA 10% AF	10,68	1,43	13,92	389,41	1,11
PE 5% BF	10,26	4,77	15,76	475,57	1,24
PE 10% BF	7,99	4,23	12,92	424,41	0,92

Table 4. Mechanical results obtained in different formulations made with abaca and banana fiber

4 Conclusions

Banana fiber appears as another option in the vegetable fibers market, with properties very similar to those obtained for abaca fiber. Banana fiber shows better mechanical properties than other natural fibers, such as abaca, flax or hemp.

Several parts with different contents in fiber have been obtained. Fiber distribution in plastic matrix is good, obtaining good aesthetics. 10 % seems to be the maximum limit of fiber to be introduced in the rotational molding process.

Banana fiber composites show better tensile properties than abaca fiber, while results show that abaca provides better properties at flexural tests. Impact properties are drastically reduced because of the introduction of materials different to PE, as fibers or even PEMA. Furthermore, use of PEMA as coupling agent does not improve the properties of the composite.

5 Acknowledgements

The authors would like to thank Innpacto Program, as this work is part of developments achieved during BioRot Project (IPT-2011-1894-420000), funded by Ministerio de Economía y Competitividad and cofinanced

with FEDER funds, and to the PPRC staff at Queen's University of Belfast for their assistance in the tests carried out in their facilities.

References

[1] Cantero G., Arbelaiz A., Llano – Ponte R., Mondragon I. Effects of fiber treatment on wettability and mechanical behaviour of flax/polypropylene composites. *Composites Science and Technology*, **63**, pp. 1247 – 1254 (2003).

[2] Carus M., Gahle C., Pendarovski C., Vogt D., Ortmann S., Grotenhermen F., Breuer T., Müssig J., Steger J., Brockmann H. *Studie zur Markt- und Konkurrenzsituation bei Naturfasern FAOSTAT*. Agency of Renewable Resources, Federal Ministry of Food, Agriculture and Consumer Protection, FKZ 22020005, Gülzow (2008).

[3] Biagiotti J., Puglia D., Torre L., Kenny J.M. A systematic investigation on the influence of the chemical treatment of natural fibers on the properties of their polymer matrix composites. *Polymer Composites*, **25**, pp. 470 – 479 (2005).

[4] Yuan X., Easteal A. J., Bhattacharyya D. Mechanical performance of rotomoulded wollastonite-reinforced polyethylene composites. *International Journal of Modern Physics B*, **21**, pp. 1059-1066 (2007).

[5] Wang B., Panigrahi S., Tabil L., Crerar W. Pre-treatment of flax fibers for use in rotationally molded biocomposites. *Journal of reinforced plastics and composites*, **26**, pp. 447 – 463 (2007).

[6] Torres F.G., Aragon C.L. Final product testing of rotational moulded natural fiber reinforced polyethylene. *Polymer testing*, **25**, pp. 568-577 (2006).

[7] Chang W.C., Harkin-Jones E., Kearns M., McCourt, M. *Class fibre-reinforced polyethylene composites in rotational moulding* in *29th Annual Technical Conference of the Society of Plastics Engineers 2011(ANTEC 2011)*, Boston, USA (2011).

[8] Ortega Z., Benítez A.N., Monzón M.D., Hernández P.M., Angulo I., Marrero M.D. Study of Banana Fiber as Reinforcement of Polyethylene Samples Made by Compression and Injection Molding. *Journal of Biobased Materials and Bioenergy*, **4**, pp. 114 – 120 (2010).

[9] MohanRao K.M., Mohana Rao K. Extraction and tensile properties of natural fibers: Vakka, date and bamboo. *Composite structures*, **77**, pp. 288 – 295 (2007).

[10] Mishra S., Mohanty A.K., Drzal L.T., Misra M., Hinrichsen G. A review on pineapp.le leaf fibers, sisal fibers and their biocomposite. *Macromolecular Materials and Engineering*, **289**, pp. 955 – 974 (2004).

[11] Satyanarayana K.G., Guimaraes J.L., Wypych F. Studies on lignocellulosic fibers of Brazil. Part I: Source, production, morphology, properties and app.lications. *Composites Part A*, **38**, pp. 1694 – 1709 (2007).

[12] Yang H., Yan R., Chen H, Ho Lee D., Zheng C. Characteristics of hemicellulose, cellulose and lignin pyrolysis. *Fuel*, **86**, pp. 1781 – 1788 (2007).

[13] De Rosa I.M., Kenny J.M., Puglia D., Santulli C., Sarasini F. Morphological, thermal and mechanical characterization of okra (Abelmoschus esculentus) fibers as potential reinforcement in polymer composites. *Composites Science and Technology*, **70**, pp. 116-122 (2010).

[14] Hornsby P.R., Hinrichsen E., Tarverdi K. Preparation and properties of polypropylene composites reinforced with wheat and flax straw fibres. Part I Fibre characterization. *Journal of Materials Science*, **32**, pp. 443 – 449 (1997).