ISSN: 2230-9926

Available online at http://www.journalijdr.com

IJDR

International Journal of Development Research
Vol. 12, Issue, 04, pp. 54990-54994, April, 2022
https://doi.org/10.37118/ijdr.24155.04.2022

RESEARCH ARTICLE OPEN ACCESS

RECYCLED HDPE/SHORT COCONUT FIBER COMPOSITES: EFFECT OF COUPLING AGENT AND FIBER LOADING

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ARTICLE INFO

Article History:
Received 19th January, 2022
Received in revised form
13th February, 2022
Accepted 16th March, 2022
Published online 22nd April, 2022

Key Words:
Coconut fiber;
rHDPE/MAPE/Coconut Fiber Composites;
Recycled Composites.

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ABSTRACT

The growing concern regarding mitigation of the environmental impacts caused by human activities has led to the development of new technologies. The use of natural fibers as reinforcement in composite materials is an alternative to reduce environmental pollution and contribute to the reduction of these impacts. Therefore, this article reports the analysis of the effect of maleic anhydride-grafted polyethylene (MAPE) as a coupling agent and coconut fiber filler on the physical-mechanical, chemical, and morphological properties of high-density polyethylene composites produced from recycled HDPE/coconut fiber. In this study, rHDPE/MAPE/coconut fiber composites (coconut fiber content: 20 and 30% m/m) and compatibilizer (5% m/m) were prepared in a Teck Tril twin screw extruder (model DCT 20-40). After extrusion of the materials, the composites were crushed and conditioned. The characterization of these materials was carried out through measurement of MFI, density, hardness and water absorption through Fourier-transform infrared spectrometry (FTIR)and scanning electron microscopy (SEM) tests. The results indicated thatwith the addition of higher fiber contents, the hardness and water absorption increased, but the density of the materials was not associated with significant variations. The presence of MAPE improved fiber/matrix adhesion through physical interaction, as indicated by SEM, which showed no change in infrared peaks. With the addition of filler, the rHDPE/MAPE/coconut fiber composites had increased MFI. The study showed that coconut fiber and rHDPE composites can be alternatives for the sustainable reuse and recycling of these wastes.

Citation: ***Camila Gisele Araujo da Costa, Monica Cristina Celestino dos Santos, Elaine Vidal Dias Gomes Libano, Daniele Cruz Bastos and Patricia Soares da Costa Pereira.*** "Recycled HDPE/short coconut fiber composites: Effect of coupling agent and fiber loading*", International Journal of Development Research*, 12, (04), 54990-54994.

INTRODUCTION

The development of technologies that reduce the environmental impact caused by human beings has become a global trend due to the growing concern for the environment. In this context, the use of natural fibers as reinforcement in the manufacture of composite materials stands out. Natural fibers have advantages over traditional inorganic fibers, such as low cost, excellent resistance to ozone and temperature, good resistance to solvents and low density. Additionally, they are non-toxic, non-abrasive, are derived from biomass, are easily modified by chemical agents and are biodegradable. Finally, the development of polymeric composites with natural fibers contributes to the reduction of environmental pollution, as well as the creation of jobs and the use of available natural resources.

Among the natural fibers used as reinforcement in composite materials, coconut fiber stands out, an agro-industrial residue produced on a large scale in Brazil (ARAÚJO *et al*., 2020; MATEUS *et al*., 2020; LIMA *et al*., 2021; SARI *et al*., 2021; VANEEWARI *et al*., 2021). Coconut fibers are composed of cellulose, hemicellulose, lignin, pectin and minerals. When compared to other natural fibers, such as jute, sisal and banana fiber, it stands out for having a higher percentage of lignin, a component that provides greater rigidity and resistance to the fiber (KUBURI *et al.* 2017; BABJI *et al*., 2020; WEARN *et al*., 2020). In Brazil, more than 1.95 billion coconuts are produced per year, and only a small fraction of the husks are used, making it important to develop alternatives for the use of this residue (BRAINER *et al.,* 2020). The production of polymeric composites can result in economic growth combined with sustainable development.

However, compatibility with the polymer matrix can be a limiting factor, since the matrix is predominantly hydrophobic while the fibers have hydrophilic character (WEARN *et al*., 2020). This polarity difference can result in a decrease in the mechanical performance of the composite due to lack of adhesion between the phases. In order to increase the interaction, and consequently the mechanical strength of the composites, it is necessary to use a compatibilizing agent (ROSSI, 2017; LIU *et al*., 2019; SARI *et al*., 2019). High-density polyethylene (HDPE) is one of the most widely used polymers to obtain fiber-reinforced composites, due to its attractive characteristics, such as low cost, ease of processing and recycling, in addition to good impact resistance (ALMEIDA, 2015; SADIK *et al*., 2021). Thus, the development of HDPE composites with coconut fibers is of great relevance. This work aimed to investigate the effect of maleic anhydride-grafted polyethylene (MAPE) as coupling agent and coconut fiber loading on the physical-mechanical, chemical and morphological properties of recycled high-density polyethylene (rHDPE)/coconut fiber composites.

Experimental Procedures

MATERIALS

The polymeric matrix used in this work was a recycled homopolymer (recycledhigh-density polyethylene, or rHDPE),provided by the company Peterlu, located in the city of Seropédica, Rio de Janeiro State. The coconut fiber *in natura* was acquired fromEmpresaCoquim, São Paulo, SP, Brazil. The coupling agent, a functionalized one (maleic anhydride-grafted polyethylene, MAPE) was donated by Polybond, under the trade name Polybond 3009, with 1.0% maleic polyethylene (nominal) and flow rate of 4g/10min (ASTM D1238, 190°C/2.16Kg).

Coconut fiber processing: The fibers were washed in water for 1 hour with agitation at 80 °C, sieved and oven dried at 40 °C for 24 hours. Before processing the composites, the fibers were dried for 1 hour at 100°C.

Composite processing: The rHDPE/MAPE/coconut fiber composites were prepared with coconut fiber (20 and 30% m/m) and compatibilizer (5% m/m) in a Teck Tril twin screw extruder (model DCT 20-40), with zone temperatures ranging from 90 to 190ºC from the head to die, respectively,and 65 rpm rotation. After the extrusion of the materials, the extrudedmaterial was crushed and conditioned. Table 1 shows the nomenclature and the mass proportions of each component obtained.

Table 1. Composition and Nomenclature of the Materials

Nomenclature	rHDPE (% m/m)	Coconut fiber (%m/m)	MAPE (%m/m)
rHDPE	100	-	-
C20		20	-
C20Co5%		20	5
C30		30	-
C30C05%		30	5

Characterization: For characterization, the specimens for the different tests (density, hardness, FTIR, water absorption and SEM) were stamped from plates obtained by compression in a bench press (Marconi model MA 098). These plates were obtained using a temperature of 190 °C, pressure of 5 tons for 3 minutes and then they were cooled in a cold press for 3 minutes, thus obtaining the films of the materials. MFI analysis was performed with the polymer in the form of pellets.

Melt flow index (MFI), density and hardness: The materials were characterized according to the melt flow index, MFI (ASTM D1238-13), density (ASTM D792-13) and hardness (ASTMD2240–13).

FTIR: The rHDPE/MAPE/coconut composites were characterized by Fourier-transform infrared spectrometry (FTIR), with a Nicolet 6700 FTIR spectrometer (Thermo Scientific). The samples were mounted on an attenuated total reflectance (ATR) accessory equipped with ZnSe crystal prior to scanning. The spectra were obtained with accumulation of 120 scans and resolution of 4.182 cm^{-1}.

Water absorption (WA): The WA test was carried out according to ASTM D-570.Three conditionedspecimens (20 x 20 x 20 mm^3) wereplaced in a container of distilled water maintainedat a temperatureof 23±1°C, resting on theedgeand entirelyimmersed. At the end of 2 h, 24 h and 720h, the specimens were removed from the water one at a time, wiped with dry cloth to remove all surface water, andweighed immediately with an analytical balance (Marte, UX4200H model) with 0.001 g accuracy.

SEM: Scanning electron microscopy (SEM) was performed using a Jeol (Model JSN 6490LV) microscope, to observe specimens coated with gold. Cryogenically fractured cross-sections of the samples were assessed, and the images were obtained at 6 k magnification, at 30 kV.

RESULTS AND DISCUSSION

The MFI values (Figure 1) showed that the fluidity of thethermoplasticmatrix was considerably affected by theamount of fiber and coupling agent added.The addition of reinforcing filler normally decreases the MFI of composite materials. The presence of fibers in the polymer matrix reduces the mobility of the chains, restricting the flow of the molten polymer, and therefore increases the viscosity (Jesus, 2014; Bastos *et al.*, 2018).Surprisingly, the greatest resistance to flow (lowest MFI) was found for rHDPE. The flow resistance of all composite formulations decreased as the amount of coconut fiber increased. Similar results were found by Gomes *et al.*(2018), who developed cashew nutshell powder- rHDPE composites using a thermo-kinetic mixer. The C30Co5% composite showed the highest MFI(1.587 g/10min).

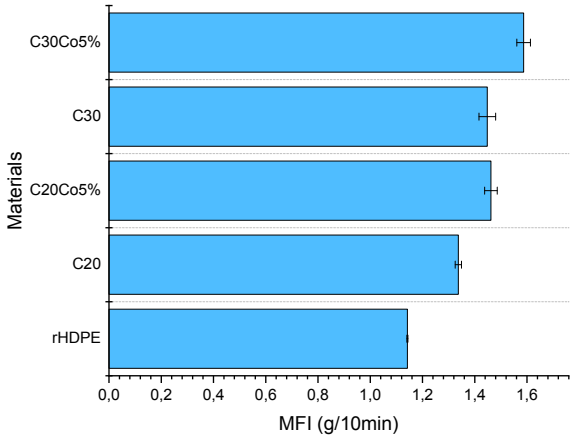

Figure 1. Results of melt flow indexof the materials (rHDPE/MAPE/coconut fiber)

Figure 2 shows the results of density analysis of the materials.The density was in the range between 0.752 and 0.798 g/cm^3. The presence of fibers and the coupling agent did not promote significant changes in the density. However, there was a tendency for increased density of composites as a function of the concentration of added fibers and with the incorporation of coupling agent. The PE-g-MA and the additives present in the recycled HDPE, which act as plasticizers, promote good matrix/filler adhesion. Similar results were found by Choi *et al.* (2006), who developed rice husk–polystyrene composites, and by Fletes*et al.* (2021) for rHDPE-maple wood fibers with and without surface treatment.The C20C05% composite showed the highest density (0.798g/cm^3). The hardness values (Figure 3) showed that the hardness of the thermoplastic matrix was significantly affected by the amount of fiber and coupling agent added. The hardness increased after fiber addition, proportionally with its concentration. The C30 composite (62.7 Shore D) presented greater hardness than C20 composite (59.7 Shore D).

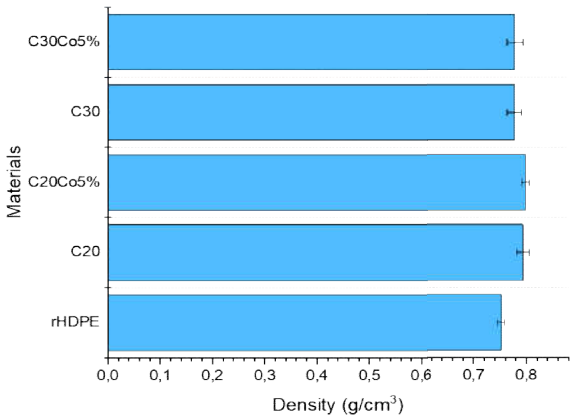

Figure 2. Density results for materials (rHDPE/MAPE/coconut fiber)

The results obtained by Durowaye*et al.* (2014) revealed a progressive rise in hardness from 0 to 20% in a composite consisting of coconut shell and palm fruit fibers with polyester. Talabi*et al.* (2012) also reported increased Rockwell hardness of a polymer matrix reinforced with coconut shell powder. After adding the coupling agent, the C20C05% composite showed the highest hardness (66.4 Shore D). There was an increase of approximately 17% in relation to the rHDPE matrix, suggesting that a possible interaction was taking place between the rHDPE and coconut fiber, favored by the addition of the coupling agent.

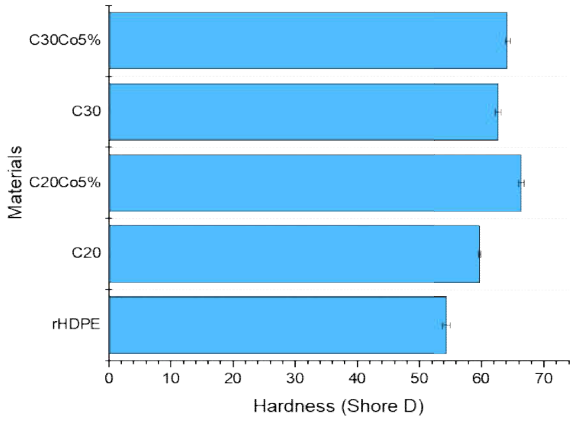

Figure 3. Hardness results of materials (rHDPE/MAPE/coconut fiber)

The water absorption (WA) properties of the rHDPE/MAPE/coconut fiber composites with or without addition of the coupling agent were analyzed through WA measurements. The results are shown in Table 2 and Figure 4. The high hydrophobicity of polyolefins explains the low hygroscopic power of rHDPE, which had very low WA, ranging from -1.852 to 0.405%. On the contrary, the WA of rHDPE composites was significantly increased after fiber addition, proportionally with its concentration. The composites without coupling agent (C20 and C30) had higher WA values, ranging from 8.570 to 13.302%, probably due to the presence of fibers and the weak fiber/polymer interaction. The composite with coupling agent (C205% and C30Co5%) had WA variation from 6.153 to 10.48%. The addition of the coupling agent (PE-g-MA) in the thermoplastic matrix promoted lower WA values. The composite with the lowest fiber and compatibilizer contentshad the lowest WA (C20Co5%). Similar results were found by Satapathy*et al.* (2015). They reported that with the addition of coupling agent (PE-g-MA) in rHDPE/coconut composites, the WA values declined. The coupling agent improved the interaction, promoting a reduction in WA.According to the literature, polyethylene (PE) or polypropylene (PP) grafted with maleic anhydride (MA) (MAPP) is one of the most suitable processing aids available for use in PE or PP composites reinforced with natural fibers (Rahayu*et al.*, 2020; Pak *et al.*, 2018; Gao *et al.*, 2012).

Table 2. Results of the water absorption test

	Conditioning time and temperature	Immersion time (h)	Averagehumidity percentage (%)
rHDPE	2hours e 23°C	2	0,174
	24 hours e 23°C	24	-1,852
	720 hours e 23°C	720	0,405
C20	2 hours e 23°C	2	8,570
	24 hours e 23°C	24	8,577
	720 hours e 23°C	720	11,795
C20Co5%	2 hours e 23°C	2	6,153
	24 hours e 23°C	24	8,871
	720 hours e 23°C	720	9,455
C30	2 hours e 23°C	2	7,629
	24 hours e 23°C	24	11,157
	720 hours e 23°C	720	13,302
C30C05%	2 hours e 23°C	2	7,629
	24 hours e 23°C	24	9,948
	720 hours e 23°C	720	10,484

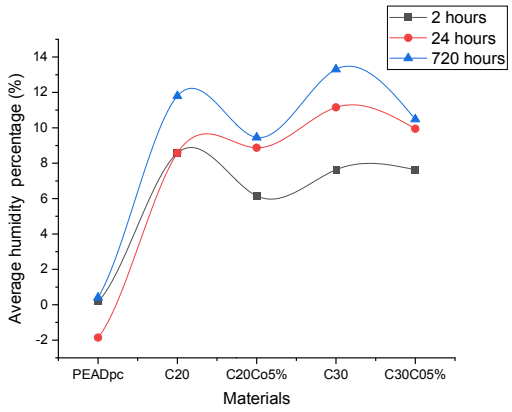

Figure 4. Water absorption results for materials (rHDPE/MAPE/coconut fiber)

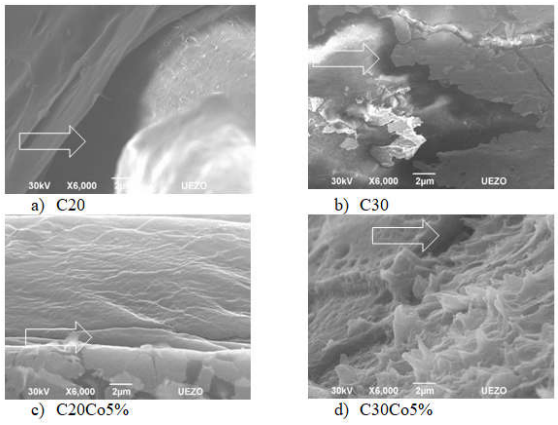

Figure 5. SEM micrographs of the rHDPE/MAPE/coconut fiber composites

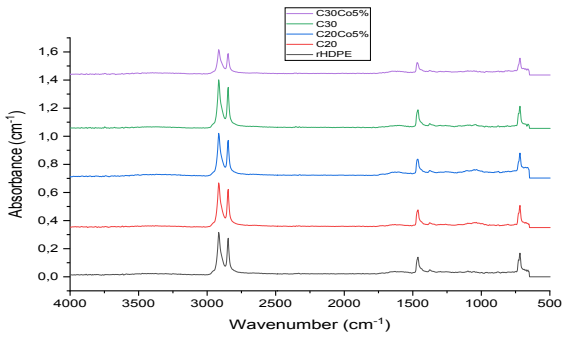

Figure 6. ATR-FTIR spectra of materials (rHDPE/MAPE/coconut fiber)

The addition of coupling and processing agents improves the fiber/matrix interaction, homogeneity, and dispersion of fibers inside the matrix, achieving better properties of composites (Liu *et al.*, 2019). The fractured surface of the rHDPE/MAPE/coconut fiber composites are presented in Figure 5. Without coupling agent, there was obvious separation between the rHDPE and fibers, as shown in Figures 5a and b (indicated by arrows), because of the incompatibility between hydrophobic matrix and hydrophilic fibers. For the composites rHDPE/MAPE/coconut (C20Co5% and C30Co5%), Figure 5 (d, e), the presence of 5 %wt of MAPE significantly improved the fiber/matrix adhesion (interface indicated by arrows). Similar results were found by Lei *et al.* (2007) and Wang *et al.* (2014). Figure 6 shows the ATR-FTIR spectra of rHDPE and the composites. Polyethylene peaks corresponding to CH, CH_2 and CH_3 appear at 2923 cm^{-1} and the peak at 1475 cm^{-1} refers to angular deformations of CH_3 (Koenig and Sypens, 2002). HDPE also exhibited C-C bending, which is normally observed with absorption bands in the 1000-1250 cm^{-1} region. Some other bands could be attributed to the additives used to improve the flow of post-consumer polymer during reprocessing (Bastos *et al.*, 2018; Torres *et al.*, 2010; Gerado *et al.*, 2020; Coelho *et al*., 2021).

Conclusion

Recycled high-density polyethylene (HDPE) was blended with differentweight percentages of short coconut fibers, with or without the couplingagent maleic anhydride-grafted polyethylene (MAPE).The rHDPE/MAPE/coconut fiber compositesshowed an increase in MFI with filler addition. The density of materials did not vary significantly. The hardness of the composites increased in the presence of higher fiber contents and in the presence of the coupling agent. The water absorption property of all composites increased with the addition of fiber. The presence of MAPE improved the fiber/matrix adhesion for HDPE/MAPE/coconut composites, as observed by SEM. The rHDPE/MAPE/coconut interface was physical, since there were no changes in the infrared peak. The results obtained showed that composites based on coconut fibers combined with recycled rHDPE can be considered good alternatives for the efficient recycling and reuse of these wastes in order to achieve sustainable products.

Acknowledgements

We thank FAPERJ (Rio de Janeiro State Research Foundation) and Peterlu.

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