Date-Palm Fiber as a Reinforcement Filler in Polymer Composites

Tariq Syed, Abdul Salam Thelakkadan, Saad Al-Hussain

SPADC-SABIC, Riyadh Techno Valley, Prince Turki Rd, PO Box 5101, Riyadh 11422, Saudi Arabia Email: Tariq.Syed@SABIC.com

Received: 9 November 2020; Accepted: 10 December 2020; Available online: 20 December 2020

Abstract: Natural fibers offer a great advantage of being used as a reinforcement in polymer matrix composites because of the many advantages natural fibers offer over conventional reinforcement fillers. Date palm fiber is one of the most available natural fibrous materials in the Middle Eastern region to be exploited as a fiber reinforcement in polymers. In the present work, the fibers extracted from the date palm tree trunk, branches, and leaves were used for the reinforcement of the polypropylene matrix. Electron microscopic images show excellent bonding between the fiber and matrix as no fiber pullout is observed. The thermal (heat deflection temperature) and mechanical properties (Izod impact, tensile and flexural modulus) of the composites increased with an increase in the fiber loading from 20% to 60%, which in turn resulted in excellent mechanical properties in the final product. The work has immense significance in using date palm as an easily available natural resource for a useful product. **Keywords**: Date palm; Composites; Reinforcement; Polypropylene; Mechanical properties.

1. Introduction

Fillers are used in polymers for a variety of reasons. Fillers reinforce the polymers by imparting additional functionalities to the polymer-like improved modulus, color, and cost reduction [1]. Common fillers used in polymers include natural or synthetic inorganic or organic materials. Talc, Mica, Calcium carbonates are examples of some common inorganic particulate fillers commonly used in polymers, while fibrous materials such as glass fiber, carbon fiber aramid fiber, and natural fibers are commonly used as fibrous reinforcement additives [2-5]. These fillers/reinforcements, whether particulate or fibrous, and most of them are inorganics, are used in polymers to increase the stiffness, hardness, and to reduce the cost [6]. Due to the inherent poor compatibility between generally hydrophobic polymer matrix and most of the fillers, several types of treatments are required to improve the interfacial bonding between the matrix and the reinforcement filler [7]. If the filler is not chemically bonded to the matrix, the enhancement in the desired properties will be poor in terms of internal strains, porosity, environmental degradation, moisture absorption, and mechanical properties. Due to many advantages over common fibrous fillers like glass fibers, natural fibers present advantages such as low cost, excellent resistance to solvents, and temperature. In addition, they are non-toxic, non-abrasive, and easily modified by chemicals. In a desert region, natural resources are scarcely available, especially water and wood. For that reason, alone it puts tremendous pressure to use or recycle whatever little natural resources are available.

Date-palm (DP) tree fiber is the most abundant naturally occurring fiber in the desert region of the Arabian Peninsula (see Fig1). Currently, date palm tree fibers are either used as mulch or burned to avoid insects and space recovery. This natural fiber is a good candidate to be used as a reinforcement in the polymer matrix [8-9]. Thus, the composite of date palm fiber with polyolefin, like any other composite, reduces the material consumption, and lowers the cost of the product, and is an efficient way of reusing the date palm tree. Therefore, it is a green solution with a lower CO2 footprint. Many studies have already been carried out by using fibers from either date palm leaves, trunk, etc. (cellulose-based fiber only). In all the aforementioned works, the palm fibers are chemically treated before they are used as reinforcement in polymers to make sure that there is good adhesion of the fiber to the polymer matrix. The chemical treatment of the fiber makes the final product very expensive mainly because of the very low fiber yield, use of only selective parts of the tree for fiber, which all makes the material commercialization almost impossible. The tedious process of fiber selection includes segregation by type (sheath, leave, trunk, etc.), extraction of cellulosic fibers and removal of lignin, selection of proper size (length), and finally chemical treatment [10]. Exclusive fiber treatments were employed, which involves chemicals that are an extra processing step [11 -12]. This results in a very poor yield of date-palm fiber.

The non-polarity of polyolefin makes it difficult for using with other polymers, additives, and fillers. Compatibilizers and coupling agents such as silanes, maleic anhydride-modified polyolefin, and Zirconates are hence, used to homogenize the mix and to facilitate good fiber-matrix bonding [14-17]. Maleated PP (e.g.

MAHgPP, MAH-SEBS) is commonly used as a coupling agent for natural fiber-based composites. These coupling agents form covalent/hydrogen bonds with the hydroxyl groups on the fiber surface and thus enhance the muchneeded bonding between the fiber and the matrix [14-16]. Impact modifiers, usually elastomeric or olefinic based are also used to improve impact resistance/toughness of the composites [17]. Studies were carried out using coupling agents at high loading levels and using date-palm fiber from tree branches. The fibers were chemically treated (which removes lignin from the sample and retain cellulosic fiber material only). Then it was washed and dried in an oven. Polypropylene is one of the most common polymers used in making composites because of its exceptional properties like high strength, stiffness, good chemical resistance, and weldability, which render it applicable in several corrosion-resistant structures [18]. The compound was formulated and hand mixed. Lumps were produced by an extruder. It was then granulated and test specimens were injection molded. Adhesion between the fiber and matrix was reported as good [19-20].

The good bonding between the matrix and fiber helps in stress transfer from the matrix to fibers to achieve favorable properties in the composites. If only naturally compatible fibers and matrices, such as cellulose fiber and cellulose ester polymers are used then there is not much need for any compatibilizers and coupling agents. The objective of the current study is to use date-palm fiber from a whole tree, not just selected parts of the tree, as against the previously reported studies. This makes a hundred percent material available contrary to thirty percent or even less when specific parts of the tree or only cellulosic material were segregated and used. Using fiber from the whole tree not only saves material and elevates supply chain position but also saves time, which is required to prepare the usable fiber. The whole tree is fed to a shredder and fibers are obtained. The very large or very small size of the fibers was removed through screening. The remaining fibers, 4-10mm long were collected. Impinged dust in the material from the desert environment over the years are removed by simply washing with tap water. No chemical treatment was used therefore, it is safer for the environment and no lignin/ cellulose separations occur either, thus, utilizing a hundred percent of the much scarce resources of the deserts, water, and wood. We have also tried higher fiber loading than what appeared in the literature, to increase the feasibility of the product. An optimal level of different ingredients was used in the formulation. The end product was characterized by mechanical, thermal, and matrix-fiber bonding. In the future, other functional additives can be incorporated for color, weathering resistance, flammability resistance, etc. The composite is useful in making a fence, facades, lumber, wood-like decorative articles, etc. Thus, recycling and conserving natural resources, which is an indispensable necessity of the dessert region, can be realized.

2. Experimental

A homo polypropylene (high melt flow: MFR = 47 g/10 min @ 230° C & 2.16kg load) was used as a matrix, maleic anhydride grafted PP was used as a coupling agent. Impact modifier, lubricant, thermal and processing aids were also used in the formulation before compounding in a twin-screw extruder. 4 - 10 mm long date-palm fibers (see Figure 1) were prepared by shredding the whole date palm tree, screened for bigger particles, and finally washed with water and dried. The dried date palm fibers are used as such for compounding. The fibers were then introduced into the polymer at a suitable location upstream in the extruder via a side feeder, which in turn was fed via gravimetric feeder. The rest of the ingredients were tumble-mixed and fed in the extruder's throat via gravimetric feeder. The extruder processing temperature was kept between 180-200°C. Composite strands coming out of the extruder were cooled in a water bath and pelletized into pellets.

Grade	Ingredients	PPDPF20	PPDPF40	PPDPF60
		(%)	(%)	(%)
PP579S	PP-Homo	63.4	43.4	23.24
DPF	Date-Palm Fiber	20	40	60
CA100	MAHgPP	3	3	3
30BA402	Ethylene-Butyl Acrylate Copolymer	10	10	10
Irgafos168	Phosphite Processing Stabilizer	0.08	0.08	0.08
Irganox1076	Phenolic Antioxidant	0.02	0.02	0.02
Calcium Stearate	Calcium Octadecanoate	0.5	0.5	0.5
Talc	Talc	3	3	3

1 abic 1.1 ormanations abou for making the composites with date paint not

The pellets were injection molded to obtain ISO test specimens (Figure 2). Thermal and mechanical properties and electron microscopy were carried out on the samples to characterize and evaluate the composite.

3. Results and discussions

The processing temperature during compounding and molding was kept below 200°C throughout the processing.

When the processing temperature during injection molding was raised above 200°C, the fiber undergoes degradation and burning which was obvious from the smell and the shift in color (see Figure 3). The degradation of the fibers was also verified by Thermogravimetric analysis (TGA) and it was found that the degradation and burning start around 230°C (Figure 4). Therefore, the melt conversion process should be kept well below 200°C. No effect of date-palm fiber loading was observed on %crystallinity of the PP, which was between 34-38% according to the DSC test (Figure 5). Therefore, it appears that the modulus of the composite depends on fiber loading percentage and adhesion of fiber to the polymer matrix.



Figure 1. Date-Palm tree fiber (4-10mm)



Figure 2. Injection molded ISO test specimens. Composite samples containing 20%, 40% and 60% of date palm fibers.

Nice woody color and texture	color turned dark gray due to burned wood
The second s	
	a faith and a started

Figure 3. Effect of processing temperature on the degradation behavior date palm fiber

After molding, the fibers in the finished product were characterized by SEM (see Table 2). The final fiber length after extrusion and injection molding is between 3 to 4mm, which is a typical length of reinforcements in the composite. Thus, it points that the extrusion and injection molding were done at optimal conditions.

Further evaluation by SEM shows that the fiber has a rough surface, the diameters vary from 95 to 500 micron,

and that the fibers are not distributed uniformly (Figure 6, Figure 7). Optimizing the extruder screw design will further help in improving the dispersive mixing of fiber in the PP matrix.



Figure 4. TGA thermogram of date-palm filled PP, showing the onset of fiber degradation at 230°C



Figure 5. DSC thermogram of ate-palm filled PP

Table 2. Date-Palm Fiber Characterization by SEM after compounding in three samples

			<u>.</u>
Mean (mm)	SD (mm)	Min (mm)	Max (mm)
1.17	0.822	0.13	3.49
0.29	0.224	0.02	1.13
1.32	0.661	0.15	3.11
0.32	0.206	0.06	1.01
1.31	0.708	0.06	4.01
0.26	0.197	0.03	1.01
	Mean (mm) 1.17 0.29 1.32 0.32 1.31 0.26	Mean (mm)SD (mm)1.170.8220.290.2241.320.6610.320.2061.310.7080.260.197	Mean (mm)SD (mm)Min (mm)1.170.8220.130.290.2240.021.320.6610.150.320.2060.061.310.7080.060.260.1970.03



Figure 6. General light image of surface (1mic bar=5mm)



Figure 7. Fiber diameter: Close up cross-sectional SEM image. Part fracture appears ductile and the adhesion between fiber and matrix appears very good (Figure 8, Figure 9).



Figure 8. SEM close-up of fractured sheet, individual fiber



Figure 9. Close up SEM image with good fiber adhesion in the matrix

As shown in Figure 10, the composite exhibits better impact properties for all the fiber loading tested. As the loading of fiber increase in the composite the impact increases almost linearly. At all loading of date-palm fiber, the notched impact increased. The percent change in the notched IZOD test is indicative of good bonding between the fiber and the polymer matrix. At 60% loading of the date-palm fiber, the percentage change is 170%, which is highly favorable.

Another advantage observed by the reinforcement of PP by date palm fiber is the increase in the modulus as a function of percent loading of the fiber (Figure 11). Especially at 60% loading of the fiber, the percent change is almost doubled. Thus, good mechanical integrity in the composite was achieved which is apparent from the Flexural modus data below.



Figure 10. N-Izod impact behavior of the PP-date palm composite as a function of fiber loading.



Figure 11. Flexural modulus versus the fiber loading in the PP-date palm fiber composites

In the case of tensile modulus, an increase was observed only at a high loading of fiber that is at 60% (Figure 12). These could be the result of the impact modifier used. Impact modifiers are known to lower modulus and increase the toughness, which was observed earlier. It could also be due to the wide variation in the aspect ratio of the natural date-palm fibers used (see Figure 7).

Heat deflection temperature (HDT) measured at 1.8 and 0.45MPa (Figure 13, Figure 14) for the composite samples shows that the HDT increases gradually with increases in the fiber loading from 20% to 60% compared to the base resin.

Properties represented in the previous sections show that with higher fiber loading, the properties of the composite are improved to a great extent. This increase in the properties is attributed to the good adhesion between the fiber and the matrix as was concluded earlier by SEM.



Figure 12. Tensile modulus properties of the composites



Figure 13. Heat Deflection Temperature (HDT) of the composites under 1.8MPa load



Figure 14. Thermal property under 0.45MPa load

4. Conclusion

An optimized PP composite formulation with different loading of date-palm fiber obtained from the whole tree along with different functional additives was used in the reported work. Without segregation of the fiber based on part of the tree, cellulose or lignin content, exact sizes, and chemical treatments, improved fiber yield drastically. Excellent bonding between fiber and matrix was achieved, which in turn resulted in good mechanical properties in the product. Additives were used at an optimum level, which kept the properties and prices in check. Thus, a viable composite was created for commercial use. Other additives for various additional functionality can be incorporated, such as flame-retardants, and UV additives for weather resistance, etc. Similarly, using PP with high molecular weight or other polymer-based resins the properties of the composite can be tailored as required. No chemical treatment on date-palm fiber was used, which saves money, time, and the environment. A wide range of fiber lengths can be used, which further saves the cost of screening and rejecting unwanted sizes, which drops percent yield, but we can still optimize shredding to obtain uniform fibers to improve the properties. An optimum process was used to obtain a viable product and to show that the composite can be produced on a commercial scale rather than just a laboratory endeavor.

5. References

- [1] Xanthos, M and Todd, D.B. Kirk-Othmer, Encyclopedia of Chemical Technology. 4th Ed. New York: Wiley & Sons, Inc., 1996. pp. 290-316.
- [2] Ceresana, Market Study: Fillers (3rd edition); 2014.
- [3] Xanthos. M, Functional Fillers for Plastics. Wenham: WILEY-VCH; 2010.
- [4] Herman F Mark. Encyclopedia of Polymer Science and Technology, 4th Edition. John Wiley & Sons, Inc.
- [5] Duluth. M.N, Plastics Compounding Red Book. Edgell Communications, 2001. Vol. 24.
- [6] Rosato, D.V. Rosato's Plastics Encyclopedia. HANSER Publisher; 1993.
- [7] Vogel, S. Cats' Paws and Catapults. New York: W.W.Norton & Co; 1998. p.123-124.

- [8] Saba N, Alothman O, Almutairi Z, Jawaid M, Ghori WJ. Date palm reinforced epoxy composites: Tensile, impact and morphological properties. J. Mater. Res. Technol. 2019; 8: 3959–3969.
- [9] Oushabi A, Sair S, Hassani FO, Abboud Y, Tanane O, El Bouari A. The effect of alkali treatment on mechanical, morphological and thermal properties of date palm fibers (DPFs): Study of the interface of DPF– Polyurethane composite. S. Afr. J. Chem. Eng. 2017; 23: 116–123.
- [10] Hindi SS, Ahmed A B, El-Feel. Physico-Chemical Characterization of Some Saudi Lignocellulosic Natural Resources and their Suitability for Fiber Production., Meteorology, Environment and Arid Land Agriculture Sciences Journal. 2010; 21(2): 45-55.
- [11] Binhussain, M.A., et al. Plastic and Palm Leaves Waste Recycling A promising economic option in the Kingdom of Saudi arabia. Riyadh: King Abdulaziz City for Science and Technology (KACST).
- [12] Abu Sharkh B.F, Hamid H. Degradation study of date palm fibre/polypropylene composites in natural and artificial weathering: Mechanical and thermal analysis. Polymer Degradation and Stability. 2004; 85(3): 967-973.
- [13] Luo X, Benson R, Kit M Kevin, Dever M. Kudzu fibre reinforced polypropylene composites. Journal of Applied Polymer Science. 2002; 85(9): 1961-1969.
- [14] Lu J.Z, Wu Q, McNabb HS. Chemical coupling in wood fiber and polymer composites: A review of coupling agents and treatment. Wood Fiber and Science. 2000; 32 (1): 88-104.
- [15] Rozman, H.D, Peng, G.B and Ishak, Z.A. The Effect of Compounding Technique on the Mechanical Properties of Oil Palm Empty Fruit Bunch-Polypropylene Composite. Journal of Applied Polymer Science. 1998; 70: 2647-2655.
- [16] Quillin D, Yin M, Koutsky JA, Caulfield DF. Crystallinity in the polypropylene/cellulose system II Crystallization Kinetics. Journal of Applied Polymer Science. 1994; 52 (5): 605-615.
- [17] Sombatsompop N, Yotinwattanakumtorn C and Thongpin C. Influence of type and concentration of maleic anhydride grafted polypropylene and impact modifiers on mechanical properties of PP/wood sawdust composite. Journal of Applied Polymer Science. 2005; 97 (2): 475-484.
- [18] Alotaibi, M, Alothman, O, Alrashed M, Anis A, Naveen J, Jawaid, M. Characterization of Date Palm Fiber-Reinforced different Polypropylene Matrices. Polymers. 2020; 12: 597.
- [19] Abu Sharkh BF, Kahraman R, Abbasi SH, Hussein A. Effect of Epolene E-43 as Compatibilizer on the Mechanical Properties of Palm Fiber-Poly(Propylene) Composites. Journal of Applied Polymer Science. 2004; 92 (4): 2581-2592.
- [20] Kahraman R, Abbasi, SH, Abu Sharkh, B. Influence of Epolene G-3003 as a coupling Agent on the Mechanical Behavior of Palm Fiber-Polypropylene Composites. International Journal of Polymeric Materials and Polymeric Biomaterials. 2005; 54(6): 483-503.



© 2020 by the author(s). This work is licensed under a <u>Creative Commons Attribution 4.0</u> <u>International License</u> (http://creativecommons.org/licenses/by/4.0/). Authors retain copyright of their work, with first publication rights granted to Tech Reviews Ltd.