Flax Fibre Reinforced Poly Lactic Acid Composites by Solvent-Casting Method

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INTRODUCTION

Natural fibre reinforced composites have less processing costs, lower weight per unit volume, high abrasion resistance and are biodegradable and recyclable[1]. In addition, they have high tensile strength, high flexural rigidity and high resistance to corrosion [2]. PLA is receiving more attention among most thermoplastics [3]. This is due to the aesthetics of its products, acceptable strength and adaptability to different processing techniques [4]. However, PLA is very brittle and has poor heat deflection temperature (HDT) when used alone; these defects are improved by using short-fiber fillers [5].

Lamination methods of processing have not been very successful due to poor adhesion. We hereby report a solvent casting method which is subsequently compression molded using 1% mandelic and benzelic acids, dicumyl peroxide and zein as compatibilizers. Some physico- mechanical and thermal properties of the composites were tested.

Results

Water uptake and Gelcontent

Water uptake an index of the absorption of moisture of composites indicates crevices through which the water molecules access the interior of the composite where the more hydrophilic fiber is situated [6]. This can limit its shelf–life and eventually making it more biodegradable.

Table 1: The water uptake and Gelcontent of the samples

Composite Volume Fraction Vm	% Water Uptake	%Gel content
P	1.858	0
PF	9.011	6.537224
PF-M	10.105	6.276551
PF-B	9.241	3.204513
PF-Z	10.204	11.36251
PF-D	7.358	12.33553

Both zein and mandelic acid have hydrophilic groups in their structures which is evident in the water absorption. DCP believed to be a good crosslinking agent led to limited water imbibitions by cross-linkings. This behavior of DCP is displayed by its highest gelcontent in Table 1.

Tensile Properties

Several workers have reported decreased strength with fiber incorporation and increase in loading with respect to PLA short fiber composites [7]. They ascribed this phenomenon to the intrinsically high strength of PLA

Table 2: The mechanical properties of the composites

Sample	Tensile Properties		Flexural Properties		Impact Energy (J/m)
	Tensile Strength (MPa)	Modulus (GPa)	Flexural Strength (MPa)	Flexural Modulus (GPa)	
P	5.93	-	x	x	x
PF	30.47	1.91	24.96	2.13	59.5
PF-M	14.73	1.57	16.09	1.77	66.9
PF-B	25.68	1.62	x	x	x
PF-Z	27.47	1.89	x	x	x
PF-D	27.68	2.00	21.75	1.82	34.8

As shown in Table 2, the uncompatibilized sample recorded the highest tensile strength. Other than DCP with respect to all the tensile properties, it appeared that the solvent –casting method used for the fabrication of the composites has created favorable condition for enough fiber-matrix interface adhesion. The Flexural strength of the only fibre reinforced composite is better than either the DCP or the Mandelic acid compatibilized ones.

However, the impact test showed that the mandelic acid compatibilized sample is superior. This result is in agreement with the crystallinity result shown under the DSC result where PF-M has the highest crystallinity index; Oyama in a recent publication reported increase of impact energy of PLA with increasing crystallinity [8].

Dynamic Mechanical Analysis Result

Table 3 gave results of the dynamic mechanical analysis of the composite samples. The neat PLA recorded a storage modulus of 0.348 GPa at 30^{o}C, when compared with the fiber reinforced composite without any compatibilizers, a reinforcement factor of about 1593 % was achieved. It has been explained that characteristically PLA exhibits glassy, glass-transition, cold crystallization and liquid-flow behaviors [9].

Table 3: Storage modulus, Tg and reinforcement data of the samples

Sample	T_g	Storage Modulus GPa at 30^{o}C	Reinforcement Achieved (%)
PLA	38	0.348	0
PFI	52	5.89	1592.529
PFM	50	6.77	1845.402
PFB	52	2.41	592.5287
PFZ	54	4.9	1308.046
PFD	50	3.48	900

Thermal Properties of the Samples

Table 4: DSC data of the samples

Sample	Crystallization		Melting		%Crystallinity
	Tc	ΔH	Tm	ΔH	
P	87.66	-7.96	147.41	15	8.56
PF	97.6	-10.79	151.97	11.15	11.60
PF-M	92-36	-14.4	151.14	13.94	15.48
PF-B	91.7	-12.18	151.1	12.74	13.10
PF-Z	96.9	-7.7	152.11	12.63	8.28
PF-D	99.59	-6.04	148	7.4	6.49

Yang et al reported decrease of crystallinity with increase in cross-linking in PLA, and shifting of Tcc to higher temperatures for the highly cross-linked samples, as opposed to the slightly cross-linked ones. It is similarly observed with the composites being reported; Tcc for the neat PLA is 87^{o}C, the DCP composite indicated 100^{o}C in the extreme while the others ranged between 92-98^{o}C.

Table 5: Thermogravimetric Analysis Result

Sample	Second Curve			%Wt loss at 150^{o}C	Char Yield at 600^{o}C (%)
	Tons ^{o}C	Tmax ^{o}C	Tfin ^{o}C		
PLA	317	370	386	3.3	8.95
PFI	350	382	407	2.65	13.42
PFM	339	377	404	2.46	11.84
PFB	331	370	404	2.86	11.85
PFZ	353	386	414	3.38	12.28
PFD	357	385	411	2.6	12.8

Table5 indicated that all the composites have better thermogravimetric profiles than the neat PLA. Yang et al reported the increase of thermal stability of PLA with increase in cross-link density [9].

The micrograph of the neat PLA is seen to have a lot of voids, however the micrograph of the only fibre-reinforced PLA;PF small amounts of the fibers pulled out during the freeze-fracture which indicates very good interfacial adhesion between the components. Both PF-Z and PF-B samples showed more fiber pulling out of the matrix; this explained their poor mechanical showings. PF-M and PF-D samples on the other hand, showed not only fewer pull-outs but phenomenally the appearance of some microspheres.

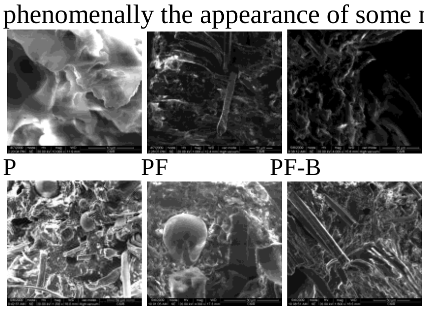

Figure 6: SEM-photo micrographs of the PLA and the composites.

They can bring about chain to chain inter-crystalline fusion of the PLA macromolecule without necessarily disconfiguring the matrix as they are few. These microspheres even though may not bring about improved adhesion but can certainly improve certain mechanical and thermal properties such as tensile and flexural moduli as well as increase crosslinking density and crystallinity. In most of these, PF-M and PF-D proved better than the fiber only composite; PF.

Conclusion

Mechanical and thermal properties of the composites have indicated that the fiber reinforcement alone without any compatibilizer did much better than the neat PLA. The mandelic acid and DCP compatibilized samples improved the flexural modulus and tensile modulus respectively. It was shown that mandelic acid led to higher crystallinity and DCP increased the crosslinking density, for both additives microspheres were seen in their SEM micrographs which indicated inter-crystalline fusion between the phases in the matrix.

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