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Enhanced Mechanical Properties and Biodegradability of Polyvinyl Alcohol and Starch Filled Water Hyacinth Fibre Biofilm

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ABSTRACT

Single-use plastic packaging that is difficult to biodegrade has been used extensively for several decades, contributing to pollution. As a result, the development of biodegradable plastic packaging has become essential to solve the ever-growing issue of disposing of plastic waste. Materials such as polyvinyl alcohol (PVA) and tapioca starch have been utilized in the creation of biodegradable biofilms. However, the addition of starch into PVA could reduce the strength and physical properties of biofilm due to the low mechanical properties of starch. Therefore, the main interest of this research is to investigate the mechanical and biodegradable properties of a biofilm composed of PVA and tapioca starch filled with water hyacinth fibres (WHF). The PVA/starch matrix was mixed with varying WHF loadings during the casting process, which produces the biofilm. Fourier Transform Infrared Spectroscopy (FTIR) analysis demonstrated a strong biofilm formation between the PVA/starch and WHF due to a good chemical interaction. Furthermore, the biofilm has become more stiff and rigid as a result of the addition of WHF, along with the increased in tensile strength and Young's modulus. However, the addition of WHF had consequently decreased the tensile strain at break of the biofilm. Because the biofilms were soluble in water, they lost weight when immersed in and stirred around in distilled water. Next, as the concentration of fibre increased, so did the biodegradability of the film. Based from the results, it can be inferred *that 10% w/w was the ideal WHF loading, producing good mechanical, biodegradable and solubility qualities. Since it is a numerous and accessible material, using water hyacinth, which is frequently thrown away because it disturbs the aquatic ecosystem, in the process of producing biofilms is a promising alternative to the packaging industry.*

Keywords: Polyvinyl Alcohol; Starch; Water Hyacinth; Biofilm; Packaging

INTRODUCTION

Material that can be broken down by the enzymes of living things is referred to as biodegradable material. Over the past 20 years, sustainable development, which contributes to environmental protection, has drawn a lot of attention. These are a result of their many benefits, which include their biodegradability, wide availability, lack of toxicological effects, and biocompatibility, particularly when compared to their synthetic counterparts [1]. In addition to being biodegradable, biopolymers also have low cost in comparison to synthetic polymers, air permeability, and the capacity to seal at low temperatures. Biopolymers, which are composed of diverse natural resources like cellulose, chitosan, starch, and different proteins originating from plants and animals, have been viewed as appealing substitutes for artificial plastic packaging materials. The packaging materials derived from biopolymers exhibit advantageous characteristics that enhance the sealed goods' quality and prolong their shelf life [2].

One of the most popular raw materials for the manufacturing of bioplastics is PVA. This synthetic polymer is transparent, non-toxic, and has a high degree of biocompatibility and biodegradability. In addition, it is a water-soluble material with strong polarity, good film-forming abilities, and numerous beneficial interactions with hydroxyl groups [3]. It thus exhibits the ability to combine and interact with other polar polymers. Starch is a type of material which can be produced from vegetable ingredients containing carbohydrates, for example tubers and cereals. Due to its biodegradability, low costs and abundant resources, starch-based bioplastics are gaining a lot of attention. Conversely, the fragile nature of starch, particularly tapioca starch leads to its weak physical properties. To address this drawback, a plasticizer is added to the starch, which can lessen the material's fragility and enhance its functionality [4]. Furthermore, starch has a low thermal stability and a higher absorption of moisture. Adding cellulose fibres to starch-based composites is an effective way to boost their performance. This is because, a strong hydrogen bond can form between cellulose fibre and starch due to their hydrophilic properties. Furthermore, the incorporation of smaller fibres create larger surface area, hence improves bonding between the fibres and the matrix material. This stronger bond enhances the composite's strength, durability, and overall performance. Additionally, finer fibres spread more evenly within the material, reducing gaps and making the composite more uniform and resistant to damage [5, 6].

The water hyacinth, or *Eichhornia crassipes*, is a naturally occurring plant species that floats freely in water and is considered an invasive weed. It is also one of the few natural fibres made of cellulose. This fibre material occurs naturally and has not yet reached its full potential. Due to their ability to inhibit waterway movement, obstruct irrigation channels and rivers, limit livestock access to water, reduce sunlight penetration, and alter water's pH, temperature, and oxygen content, water hyacinths are regarded as an environmental annoyance [7]. Moreover, the water hyacinth may modify aquatic organisms' habitats, diminishing the aesthetic value of streams and lowering water quality through the decomposition of plants. These plants were regarded as waste in every country because of their extreme negative effects. Consequently, scientists focused on this hyacinth plant and transformed it into a useful and aesthetically pleasing object [8]. Combinations of plant fibres can provide bio-composites that are a desirable substitute for many synthetic polymers. Because of their rapid growth and widespread availability, WHFs with a high cellulose content are a preferred reinforcing material for starch bio-composites [9].

In the production of PVA biofilm, starch plays a key role in flexibility, while the presence of WHFs improves the tensile strength. The enhanced tensile strength thereby allows the material to withstand stretching and handling, making it a suitable and eco-friendly alternative to conventional plastic packaging [10]. Biodegradable plastics can have the desired qualities if the ingredient amounts are carefully regulated. For commercial applications, starch-based polymers are frequently combined with eco-friendly thermoplastics [11]. WHFs must be separated from water and processed into short fibres in order to be used effectively. To enhance the quality of the fibre, hyacinth stem fibres undergo an additional chemical treatment [12]. Water hyacinth has been shown to have both advantages and disadvantages, so it should be used sparingly to prevent river pollution and in food packaging. Therefore, the objectives of this research are to analyse the mechanical properties and biodegradability behaviour of PVA/starch biofilm through tensile, biodegradation and solubility tests.

EXPERIMENTAL METHODOLOGY

Materials

Analytical grades of PVA (molecular weight, 115000 g/mol; degree of polymerization, 1700-1800), sodium hydroxide (NaOH) (18%) and glycerol (30%) used in this research were purchased from Acros Organics. All chemicals were used without further purification. Tapioca starch was purchased from local grocery store.

Extraction of WHF

Water hyacinth plant was harvested and cleaned to remove any dirt and pollutants. Subsequently, the leaves and roots of the water hyacinth were manually removed with scissor, resulting in a 10 - 20 mm cut to the stems. The stems were dried at 90 °C for 24 hours in the Memmert Universal oven. After that, the stems were powdered and sieved to a 200 µm size. The resulting WHF powder was then heated in 700 mL of 18 % NaOH to 80 °C for 2 hours. It was reported that the best concentration of NaOH for hemicellulose removal without compromising fibre strength is 18% [13]. After being treated, the WHF was filtered and allowed to dry further for 12 hours at 80 °C in the oven.

Preparation of PVA/Starch/WHF Biofilms

Firstly, 25 g of PVA was initially dissolved in distilled water for 30 minutes at 90 °C while being constantly stirred to ensure complete dissolution. Next, different loadings of WHF (% (w/w) from tapioca starch) were added to the matrix after the tapioca starch had been dissolved in 5% (w/v) distilled water, as shown in Table 1. Following this, 5 mL of glycerol, which serves as a plasticiser, was added, and the heating temperature was kept at 90 °C while constant stirring was set at 300 rpm. The mixture was then casted into a silicone square mould measuring 10 x 10 x 1 cm. The biofilm was finally dried for 24 hours at 40 °C in a drying oven. The biofilms were kept dry in a desiccator for 24 hours prior to being tested.

Table 1: The composition of PVA/starch/WHF biofilms.

Sample code	Glycerol (mL)	PVA (g)	Tapioca Starch (g/40mL) aquadest	WHF (% w/w from dry starch basis)
WHF0	5	25	2	0
WHF5				5
WHF10				10
WHF15				15
WHF20				20

FTIR Spectroscopy

The Perkin Elmer Frontier FTIR spectroscopy was utilized to ascertain the functional group of every sample. The test samples were in the shape of square films, each measuring 1 cm x 1 cm. With a resolution of 4 cm^{-1}, the scanning range was set between 4000 - 600 cm^{-1}.

Tensile Test

The tensile test of all biofilms sample was determined by using Instron Tensile Tester having a load cell of 5 kN. All samples were prepared and tested according to ASTM D 882. The crosshead speed during testing was set at 2mm/min at room temperature. The tensile test was carried out by five times repetition for each sample variation.

Biodegradation Test

Soil burial test method was used to determine the biodegradation of biofilm samples. The soil under investigation is the blended soil purchased from Eco-Shop. Every sample was first dried for 24 hours at 40 °C in an oven to achieve a constant weight, and then the initial weight was measured using a precision balance. The samples were consequently buried for 20 days in soil. They were then cleaned with distilled water and allowed to dry for 24 hours at 40 °C in the oven. Before being weighed, the samples were kept in a desiccator for an additional 24 hours. The final weight determination was conducted using the same precision balance until constant weight achieved. The percentage loss of all samples was determined as Eq. (1).

$$\text{Weight Loss (\%)} = \frac{(\text{Weight before Burial} - \text{Weight after Burial})}{\text{Weight after Burial}} \times 100 \quad (1)$$

Solubility Test

The film samples were cut into 2.0 cm^2 square sections, and the dried mass of the film was precisely weighed and recorded. The samples were left at 25 °C for 6 hours while submerged in 100 mL of distilled water with the agitation set to 180 rpm. After 6 hours, the remaining sections of the film were filtered. They were heated up at 110 °C to achieve a final fixed weight. Glycerol has a good water solubility range from 18% to 25%. The percentage of total soluble matter (% solubility) was then calculated as in Eq. (2).

$$\text{Water Solubility (\%)} = \frac{(\text{Initial Weight} - \text{Final Weight})}{\text{Initial Weight}} \times 100 \quad (2)$$

RESULTS AND DISCUSSION

FTIR Analysis

Figures 1 and 2 show the FTIR spectra of untreated WHF, treated WHF, PVA, PVA/starch, and PVA/starch/WHF, respectively. The three primary constituents of natural fibre were lignin, hemicellulose, and cellulose. There were three components that made up the natural fibres namely an ester, an aromatic ketone, and alcohol groups with varying amounts of oxygen in the functional groups [14]. The presence of lignin components on the peak characteristic, detected at about 1517 cm^{-1}, corresponds to the vibration of the aromatic skeleton and the carbonyl group in the untreated WHF. It is clear that the three peaks' spectrum appeared faintly following the alkalization and bleaching operations. The existence of carbonyl (C=O) linkage, one of the characteristic groups of lignin and lignin/hemicellulose present in untreated WHF, was responsible for the peak in the spectra that appeared at 1626 cm^{-1}. Because of the NaOH reaction's elimination of the hydroxyl (OH) group, the alkali treatment can weaken hydrogen bonds [15]. In comparison to the untreated fibre, this result lowers the -OH content and increases the intensity of the peak extending between 3800 and 3300 cm^{-1} [16]. The existence of OH stretching vibration of hydrogen bonding in the OH group resulted in the appearance of the domain 3750-3200 cm^{-1}. This behaviour was linked to the presence of hydrogen bonding interactions in the cellulose molecule, which became stronger following chemical treatment [17]. Aside from that, CH groups emerged as peaks in each sample at wavenumbers 2942 and 2946 cm^{-1}, indicating the existence of cellulose molecules [18]. Because of the hydrophilic character of the fibres, the spectra appeared at the wavenumber 1600-1650 cm^{-1}, which corresponds to the bending vibration of water absorption [14].

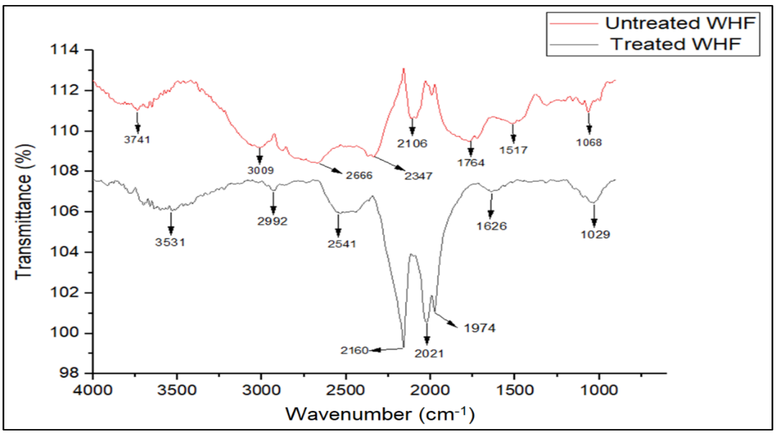

Figure 1: FTIR spectra of treated and untreated WHF.

Figure 2 shows a comparison of the chemical functional groups of PVA, starch, PVA/starch, and PVA/starch/WHF. Both PVA/starch and PVA/starch/WHF showed large peaks between 3500 and 3000 cm^{-1} wavenumber range, which correspond to the OH group of the PVA and the alkaline-treated WHF. The peak at 2946 and 2942 cm^{-1} indicates C-H stretching, while peaks of C=O groups emerged at 1029 and 1032 cm^{-1}.

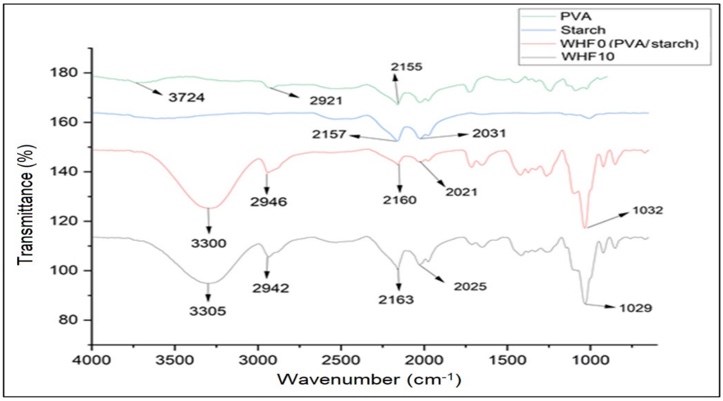

Figure 2: FTIR spectra of PVA, starch, PVA/starch and PVA/starch/WHF.

Tensile Properties

Figure 3 displays the tensile strength of the PVA/tapioca starch/WHF biofilms. It was observed that the tensile strength increased with the increased of WHF loading. The unfilled PVA/starch biofilm's tensile strength shows the lowest strength at 1.00 MPa. The highest value of 1.14 MPa was reached by the tensile strength after adding 10 % (w/w) WHF. It was reported that cellulose fibre and starch have good interaction hence contributed to the increased in strength [19]. The strength of the biofilm was, however, subsequently reduced by additional increases in WHF loading into PVA/starch. The inhomogeneous distribution of WHF in the starch matrix, may have contributed to the decrease in tensile strength relative to the increased in filler loading [20]. Figure 4 on the other hand shows the results of an investigation into the tensile strain of the PVA/starch film filled with WHF. It appears that the biofilms' tensile strain had decreased as a result of the increase in WHF. Tensile strain dropped from 129 % to 105 % when comparing unfilled biofilm of WHF0 against WHF10 with 10% WHF loading. This behaviour might have been caused by the stiffness that the cellulose structure in the WHF introduced [21]. Higher tensile strain values are preferred for packaging applications; however, this must also be proportionate to the film's capacity to tolerate the tensile stress needed to support the packing load [20].

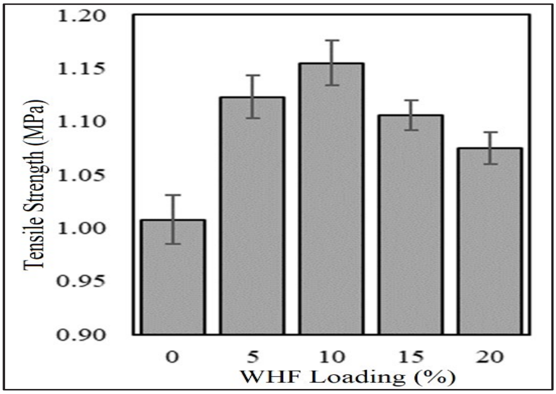

Figure 3: Tensile strength of PVA/starch biofilm filled with various WHF loadings.

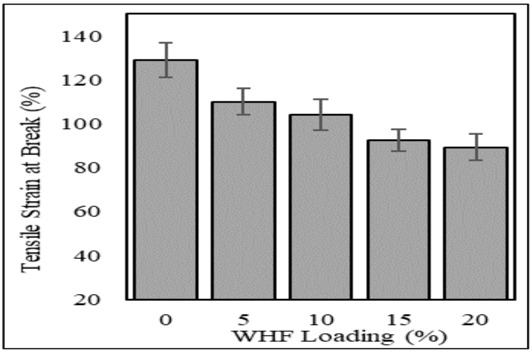

Figure 4: Tensile strain at break of PVA/starch biofilm filled with various WHF loadings.

The Young's modulus of the PVA/starch biofilms filled with WHF is shown in Figure 5. WHF20 was discovered to have the highest Young's modulus value at 4.90 MPa. A high Young's modulus is indicative of an elastic material. Given that it has the highest Young's modulus among the films, this indicates that the film with a 20% WHF loading is the most brittle. This is because WHF and tapioca starch form hydrogen bonds, which cause intermolecular interactions. The biofilms incorporated with WHF are less flexible and more rigid due to their greater Young's modulus when compared to unfilled PVA/starch biofilm. WHF's fibrous structure contributes to their tendency towards greater stiffness.

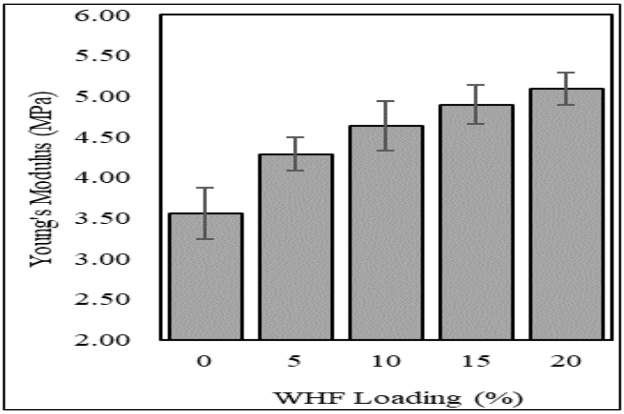

Figure 5: Young's modulus of PVA/starch filled with various WHF loadings.

Solubility

One of the most promising characteristics of the biofilms is their capacity to absorb and retain a significant amount of water, which is primarily dependent on the presence of a hydrophilic functional group in both the polymer and the monomer. Biodegradability and the solubility behaviour of the biofilm are interrelated because faster biodegradation is usually promoted by higher solubility. A biofilm that partially dissolves or absorbs water will be easier for microbial enzymes to reach, speeding up the breakdown process. Figure 6 shows the results of measuring the optimized biofilms' time-dependent (1 – 24 hours) water uptake capacity. The figure demonstrated that PVA/starch biofilm has a significantly higher water solubility rate than PVA/starch biofilm filled WHF. The OH groups presence in the polymer backbone served as the main active site for the formation of hydrogen bonds with water molecules, which is necessary for water solubility [22]. This phenomenon can be explained by the fact that the higher degree of crosslinking between the polymer chains formed the rigid and compact structure of the neat PVA/starch film, which directly affects the amorphous region where the actual solubility occurs. However, solubility behaviour rapidly declined as WHF increased and reached its lowest value of 13% for biofilms containing 20% WHF. Additionally, because of their high cellulose content, WHFs are insoluble in water. Consequently, WHF decreases the biofilm's water solubility.

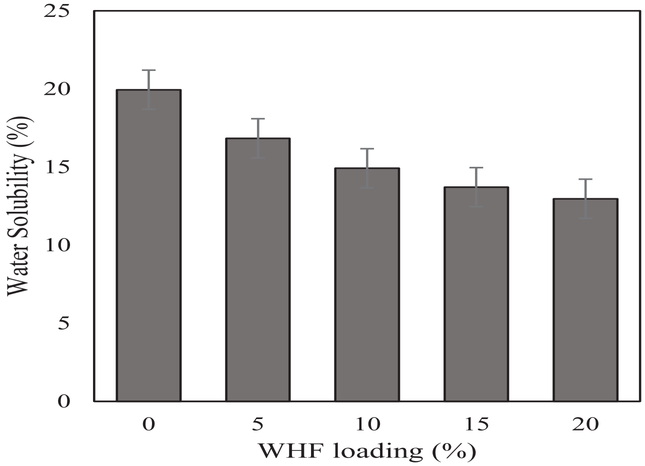

Figure 6: Percentage weight loss in PVA/starch biofilms filled with various WHF loadings as a result of solubility test.

Biodegradability

Figure 7 shows the weight loss as a percentage of biodegradation. The largest percentage of weight loss was seen in PVA/starch filled with 20WHF loadings. Despite the fact that WHF had previously reduced the biofilm's solubility, their introduction of surface roughness, microporosity, and increased surface area encouraged microbial colonisation. Consequently, the biodegradability of the resulting biofilm was greatly enhanced by the addition of WHF. Besides, natural fibres not only act as fillers but also serve as degradable components, enhancing microbial breakdown [6]. Unfilled PVA/starch film, on the other hand, degrades the least. According to reports, tapioca starch, glycerol, and PVA contain an antimicrobial that keeps microbes from breaking them down [23].

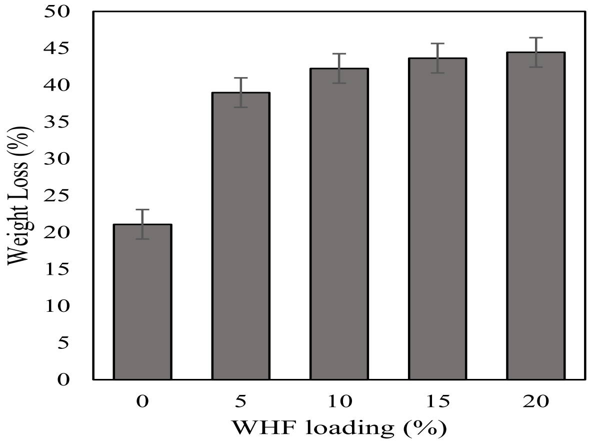

Figure 7: Percentage weight loss in PVA/starch biofilms filled with various WHF loadings as a result of biodegradation test.

CONCLUSION

In conclusion, the utilization of PVA/tapioca starch loaded with different WHF loadings yields encouraging outcomes. The composite material's capacity to form biofilms with good mechanical and barrier qualities makes it an appealing option for food packaging applications. These findings were verified by the detection of a chemical interaction between WHF and tapioca starch using FTIR. As demonstrated by the peak at 1517 cm^{-1}, the FTIR analysis verified that the PVA/starch/WHF biofilm displayed chemical interactions, indicating better intermolecular bonding that enhances the biofilm's tensile strength. Tensile strength and Young's modulus both rose as the biofilm's WHF content did. Nevertheless, additional WHF addition had decreased the biofilms' tensile strain at breakage. The addition of WHF into PVA/starch film had also contributed to an increased in biodegradation behaviour. Biofilms have a high-water solubility, therefore when submerged and stirred, the films dissolve over time. Moreover, as the WHF content increased, the biofilms' solubility decreased. The results showed that the optimum loading for the PVA/starch biofilm to produce good mechanical and physical properties was 10% w/w WHF. Therefore, WHF is essential for food packaging in order to extend its shelf life since it can increase the water resistance of PVA/tapioca starch films. However, more investigation is needed to determine their actual environmental impact. To make sure that biofilms meet sustainability objectives without unintentionally harming the environment, we should understand how they interact with it, including how quickly they break down in various environments and whether any hazardous compounds are released from them.

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