

# Bioplastics Starch-Based with Additional Fiber and Nanoparticle: Characteristics and Biodegradation Performance: A Review

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# Bioplastics Starch-Based with Additional Fiber and Nanoparticle: Characteristics and Biodegradation Performance: A Review

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## Abstract

Plastics are widely used by the community, especially as food packaging. In general, plastic raw materials are polymers which have advantages including good mechanical properties, cheap, lightweight and easy in the process of manufacture and application. However, there are still many plastic shortcomings, one of them is not easily biodegradable. Therefore, scientists and other stakeholders have shown great concern to overcome the accumulation of non-biodegradable plastics in the environment by develop plastic biodegradable. Bioplastics are one of the candidates in the highest trend because they are generally biodegradable, use natural resources and can reduce environmental pollution. Starch-based bioplastics are a popular material in the manufacture of bioplastics. However, due to it's low mechanical properties, over time natural fibers are added which are claimed to increase the strength of bioplastics and accelerate the degradation process in the soil. The addition of nanoparticles is also done quickly because the application of inorganic polymers can increase the stiffness and tensile strength of resulting bioplastic. So, in this review we describe the characteristics and biodegradation performance of several starch-based biodegradable plastics added with fibers and nanoparticles and this review expected to contribute in the future to make bioplastics which aims to take it easier for determine the material components to be used.

**Keywords** Natural fiber · Nanoparticle · Biodegradation · Food packaging

## Introduction

Plastics are widely used by the community, especially as food packaging. In general, plastic raw materials are polymers [1] which have advantages including good mechanical properties (strong), cheap, lightweight and easy in the process of manufacture and application. However, plastics still have many disadvantages, one of them is not easily degraded by the environment either by rainy or hot weather as well as microbes that live in the soil and plastic materials cannot be recycled naturally quickly [2]. The total amount of waste produced till now is around 8300 million metric tons and the resulting plastic waste is estimated at 6300 Mt, out of which only 9% has been recycled and 12% is undergoing a cineration process, the remaining 79% is still in the environment of landfills. If this cycle continues, around 12,000 million plastic waste is estimated to be collected in

environment or landfills by 2050 [3]. The solution to reducing waste buildup by burning plastic is also not the right choice, because plastic that doesn't burn completely, which is below 800 °C will form dangerous dioxins [4]. Therefore, scientists and other stakeholders have shown great concern to overcome the accumulation of non-biodegradable plastics in the environment by develop plastic biodegradable namely bioplastics [5].

Bioplastic is an innovation that has proven to be an environmental-friendly alternative that has potential to replace conventional plastics because it is biodegradable, sustainable and renewable. Bioplastics can be synthesized from natural organic materials such as polysaccharides, proteins and lipids, but starch-based bioplastics are the most promising because of their abundance in nature [6]. Starch-based bioplastics have several advantages that is biodegradable and easy to process such as potato starch [7–9], cassava starch [8, 10], corn starch [8, 10, 11], wheat starch [11], jackfruit seed starch [12] and triphala churna [13]. However, starch-based materials showed low water barrier characteristics and poor mechanical performance compared to the non-natural polymers, that is due to their high hydrophilicity and affinity to

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water, such disadvantages severely restrict their widespread application. Some ways to overcome this deficiency are mixing with other biopolymers, modifying starch, biowaste [14] and adding fiber. Natural fibers have one-dimensional and elongated structure with a round cross section. The advantages of fiber are renewability, abundance, low cost, low density, strength, stiffness, good thermal properties, and biodegradability [15]. Natural cotton, hemp, sisal, jute, flax, coconut, and sugarcane bagasse are some sources of natural fiber [16, 17].

In recent years, the incorporation of nanoparticles (NPs) into composite materials has attracted a great deal of attention for its ability to enhance the thermal, mechanical, and gaseous barrier properties of polymers [18]. Some metal oxide NPs, such as zinc oxide (ZnO) [4, 18, 21], silicon dioxide (SiO<sub>2</sub>) [7, 19], titanium dioxide (TiO<sub>2</sub>) [20, 21], calcium carbonate (CaCO<sub>3</sub>) [22], and act as antibacterial agents besides their ability to block UV radiation [18]. With this addition, composite materials containing nanoparticles can produce high-performance and innovative materials. Inorganic nanoparticles maintain excellent interfacial interactions in bioplastics and significantly enhance the mechanical properties of the matrix. According to literature, all mechanical and thermal properties of bioplastic composites are improved with the addition of inorganic nanoparticles [7]. So, this paper begins with briefly describes about classification of modified component of bioplastics. This classification aims to make a little easier for researchers to determine the bioplastic components to be made, with the hypothesis that bioplastics produced have good mechanical and biodegradability properties and renewable in nature.

## Classification of Modified Component Bioplastics

There are three types of material modifications typically used in the manufacture of bioplastics. The first is starch, then starch is added with fiber as reinforcement and the addition of nanoparticles which serves to improve the

mechanical properties of the bioplastic produced as can be seen in Tables 1, 2, and 3.

Plastics are materials that composed of molecular synthesis. It can be shaped and used as building materials such as glass, wood and metal. Their nature is adaptable to many applications using additive or alternative cross-cutting technologies. Bioplastics are a group that is divided into two qualitative criteria, namely bio-based and/or biodegradable. The term "bio-based" refers to materials derived from biomass, while "biodegradable" refers to materials which can be assimilated by microorganisms. Conventional plastics have low levels of degradation and the presence of macro and micro plastics in the soil and water, which is a threat to environment. Micro-plastics are classified as particles 1–5 mm in size [49].

Conventional plastic is a type of plastic that takes a long time to degrade. This makes conventional plastics un-eco-friendly. One solution to overcome this problem is bioplastics. Bioplastics are innovations that can prove to be great eco-friendly alternatives than conventional plastics because they are biodegradable, sustainable and renewable. There is no doubt that bioplastics are the way forward and can help reduce our dependence on conventional plastics resulting in an epidemic of plastic pollution. Bioplastics can be synthesized from natural organic materials such as polysaccharides, proteins and lipids, plus starch which is abundant in nature. Various researchers have succeeded in producing various starch-based biodegradable bioplastics [50]. Starch is a natural polymer extracted from plants and can be used to produce biodegradable plastics due to its nature, and low cost [36].

Starches from various sources differ in their chemical composition and in the structural characteristics of glucans and granules due to genetic, environmental and nutritional factors that expose plants to during their development. On the inside of starch amylose granules, lipids, phosphorylated residue, and long lateral chains of amylopectin interact between them to avoid water uptake. In contrast, the high percentage of amylopectin, especially with short lateral chains, allows hydration via freer hydrogen bonding which results in a gel that is susceptible to retrogradation

**Table 1** Starch-based bioplastics

No	Starch	Tensile strength average (MPa)	Thermal stability average (°C)	Flexural average (%)	Biodegradation average	References
1	Potato	5–25	95–140	10–80	Degradate after 5 day	[7–9, 11, 19, 22, 23]
2	Corn	1.3–35	54–267	2.1–124	81% degradate after 30 days	[8, 10, 11, 19, 20, 23–27]
3	Cassava	1–8.8	30–300	1–55	40% degradate after 7 days	[8, 10, 24, 28–36]
4	Banana	0.1–3	70–95	25–30	60% degradate after 90 days	[25, 37–41]
5	Sago	0.5–68	342–357	6–14	95% Degradate after 28 days	[42–45]
6	Rice straw	11–22	100–200	33–140	75% Degradate after 78 days	[46–49]

**Table 2** Bioplastics starch-based with additional fibers

No	Fibers	Starch	Tensile strength average (MPa)	Thermal stability average (°C)	Flexural average (%)	Biodegradation average	References
1	Pineapple microfiber	Potato	n/a	53 (3)–(–59)	n/a	n/a	[56]
		Corn	51	n/a	n/a	23 Degradate after 28 days	[57]
		Cassava	14–18	n/a	5–9	n/a	[58]
		Poly-propylene	55–80	n/a	12–16	n/a	[59]
		Polyester	36–103	30–120	n/a	n/a	[60–62, 66]
		Polyactic acid	22	57	n/a	23	[63]
2	Sisal	Corn	3–8	195–205	60	n/a	[15]
		Epoxy	25–30	n/a	2.0–2.5	n/a	[69]
		Poly-propylene	17–35	110–300	10–32	n/a	[72, 73]
		Polyactic acid	55	50–70	n/a	n/a	[74]
3	Bamboo	Cassava	15–23	n/a	15–20	23 Degradate in 15 Days	[75]
		Seaweed	47–109	n/a	14–22	n/a	[76]
		Polyactic acid	15–49	37 180	10	n/a	[77, 78]
4	Sugarcane bagasse	Cassava	1.1–2.5	n/a	7–15	n/a	[80]
		Acetylated Corn Starch	6–35	n/a	13–21	n/a	[81]
		Poly-propylene	26–31	n/a	11–16	n/a	[82]
5	Cellulose	Sago	3–10	300	n/a	50% Degradate after 5 days	[85]
6	Curaua	Corn	20–35	320–490	1.5–3	n/a	[86]

n/a not applicable

**Table 3** Bioplastics based on starch with nanoparticles

No	Nanoparticles	Starch	Tensile strength average (MPa)	Thermal stability average (°C)	Flexural average (%)	Biodegradation average	References
1	ZnO	Jeneng	1–3	361–390	2–5	30–40 days	[4]
		Carboxy-methyl Cellulose	n/a	100–150	n/a	35 days	[18]
		Cassava	4–196	300–317	5–334	7 days	[33, 51]
		Banana	35–40	n/a	5–10	90 min	[39, 87]
2	SiO <sub>2</sub>	Potato	1.2–15	100–200	55–84	5 days	[7, 88]
		Corn	0.8–1.6	120–160	55–80	40 days	[88]
3	TiO <sub>2</sub>	Corn	3–4	200–300	62–88	64% degraded after one month	[20]
4	ZnO+TiO <sub>2</sub>	Cassava	8–20	n/a	15–30	3 days	[21]
5	CaCO <sub>3</sub>	Potato	30–40	n/a	10–16	n/a	[22]
		Cassava	1–3	200–250	39	n/a	[30]

n/a not applicable

when stored for long periods. Smaller starch grains have a larger superficial area which allows fast hydration, however the surface pores and channels on the surface of starch granules increase water uptake. Thus, the higher hydration rate increases the swelling capacity, viscosity and gelatinization of the starch granules. However, the collection of chemical compounds found in starch granules and their interactions are also involved in the thermal properties of starch [51].

Amylose starches are a more efficient raw material for bioplastic synthesis than normal starches as previously established by [49]. One effective method of obtaining amylose—only starch is by modifying polysaccharides in plants by utilizing transgenic methods. In a similar study conducted by [49] transgenic barley was observed to produce starch containing 99% amylose. Thus, bio-plastics synthesized using starch from plants can prove to be a good substitute to certain flexible plastics. Presently, starch finds extensive

applications in many sectors such as agriculture, medicine, engineering, food, and packaging etc. The sources of the starch were various as can be seen in Fig. 1 and Table 1

The usability of any bioplastic depends upon their specific physiochemical properties which determine for what purpose the bioplastic is suitable. Various previous studies document and demonstrate the practical usability and applicability of different types of starch bioplastics in food packaging, shopping bags, agriculture, and medicine. It is common knowledge that the primary disadvantages of utilizing starch are its water dissolvability and low mechanical strength. Past endeavors have been made to enhance the characteristics of starch; mixing it with degradable synthetic polymers, like polyvinyl alcohol and polylactic acid, using lingo cellulosic biomass as are in forcing filler and using the agricultural waste [52]. Many previous studies have attempted to produce bioplastics from natural starches, as can be seen in Table 1

In the previous study, thermoplastic starch from three starch sources (potato, cassava and corn) and the effect of chitosan coating these composite employed by [8]. Corn starch provides a thicker outer shell and higher density compared to other composite. There is no significant difference in water absorption between three types of starch. However, corn starch obtained higher tensile strength (TS) than potato and cassava and this statement similar with [10, 11]. Whereas [9] explored the morphology and physical properties of gelatin/potato starch edible biopolymer films using casting method with varying starch content (from 0 to 50 wt%) and the addition of glycerol as a plasticizer. Microstructure morphology of films examined by optical microscopy. It is found that when starch content  $\geq 10$  wt%, the phase separation results in formation a continuous phase of gelatin matrix and a minor phase of starch micro granules is observed. It was shown that the size of the coalesced starch granules linearly correlates with the surface friction

and the water barrier properties. This indicates that the phase separation mechanism influences on these properties of the biocomposite films. The mechanical properties (such as strength and elasticity), transparency and film thickness strongly depend on the starch content; they are generally deteriorated. Yet the elasticity of biocomposites films with 10 and 20 wt% increases as compared with those of pure gelatin. It is confirmed by the thermal analysis data that the glycerol is not only a plasticizer but also a compatibilizer in the systems.

On the other hand, films from potato starch were the most amorphous and hence most transparent, because its considerably higher molecular weight and the presence of more phosphate monoesters. The overall growth rate of crystallization and the resulting crystallinity decrease with increasing molecular weight. Phosphate monoester are charged groups which can influence the crystalline arrangement during starch film preparation leading to a lower ordered structure [11]. But based on [19] show that corn starch-based bioplastics were more colorless, more transparent, less opaque and presented lower moisture content than potato starch-based bioplastics. However, the thickness of bioplastics by corn starch was higher than potato starch, but the mechanical and thermal properties remained higher for the corn starch-based bioplastics. In the biodegradability assay, although the potato starch-based bioplastic had worse physical, mechanical and thermal properties, it presented higher biodegradability than corn starch-based bioplastic. The bioplastics of potato starch were biodegraded in 5 days, and those of corn starch took almost 40 days.

Zougranan et al., was report two types of plastics using local varieties cassava and corn. These are simple bioplastics, made with starch only and composite bioplastics obtained by adding a natural ingredient extracted from the bark of the medicinal woody species *Cola cordifolia* to the starch. The biodegradability of these two types of bioplastics

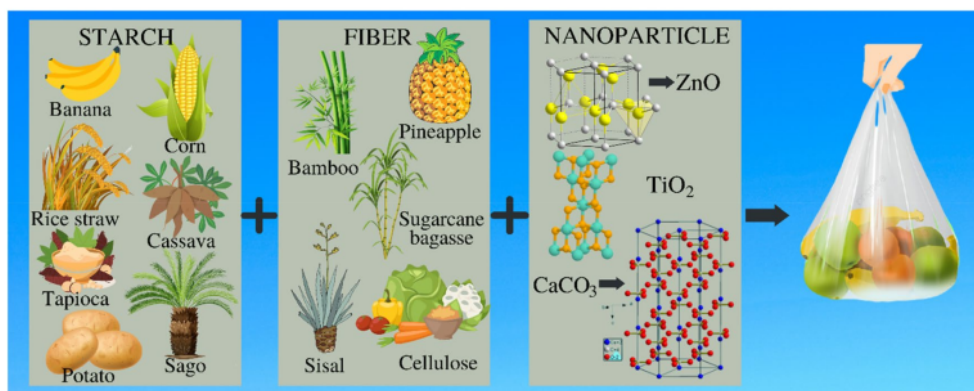


Fig. 1 Source of the starch with additional fiber and nanoparticle for increase mechanical properties of bioplastics

was assessed through burial tests in soil [24]. Cassava based bioplastics are more biodegradable than corn based bioplastics under normal study conditions. The addition of natural ingredients extracted from *Cola cordifolia* to the starting starch yields a composite bioplastic with much better biodegradability that is potentially safe for use in food packaging. Taking into account the influence of surrounding biotic and abiotic factors, it has been further demonstrated that, regardless of the bioplastic being considered, biodegradability can be significantly improved by controlling for several factors such as temperature, humidity and enrichment of microorganisms, of the burial matrix. However, the study shows that these factors can reduce biodegradability above a certain intensity limit. Above this limit, these environmental factors can inhibit the biodegradation of bioplastics. Very wet burial matrices, or rich in microorganisms with the same properties, or with high temperatures, do not promote the rapid biodegradation of simple bioplastics and composites based on corn and cassava starch. The biodegradation of starch-based bioplastics leads to acidification of the soil which can also slow down their biodegradability.

In previous study [27] structure and properties of biodegradable corn starch/chitosan composite films as affected by PVA additions was reported. The results obtained in the work demonstrate significant changes in the structure of St/Chit/PVA films with a variation of the PVA content in their composition. PVA addition of in the concentration of 10–40 wt% to the St/Chit blend enhances the intermolecular interactions between St and Chit, the higher is the PVA content the more it grows. At the same time, the phase separation between the PVA and the St/Chit matrix increases. The UV-protective properties of composite films associated with the Chit macromolecular structure are completely preserved when the PVA is added to the composition in the concentration up to 30 wt%. The transparency of the films, in this case, increases. The mechanical properties and biodegradability tests show that the PVA added to the St/Chit (70/30) matrix in an amount of 10–20 wt% is the optimal supplement providing the films with improved mechanical and functional characteristics without loss of biodegradability.

Cassava starch has also been a focus experimentation for the synthesis of biodegradable starch-based plastics. Cassava starch with the addition of some varying amounts of microcrystalline cellulose, lignocellulose nanofiber (LCNF) [29], glycerol as a plasticizer [30–32] the results of previous studies showed that the TS test of cassava starch was still very low, but if added with glycerin or polyvinyl alcohol (PVA) it would increase the strength of the bioplastic. In previous study [36] takes a closer look at the degradation of bioplastics composed from cassava starch added with glycerol as a plasticizer. The bioplastics produced were hard, smooth and transparent. Cassava starch consists of semi-crystalline structures because its granules are disrupted as

a result adding specific heat and solvents. Then, the semi-crystalline structures will turn into amorphous forms determined to be a fragile.

The mass of bioplastics buried for 6 days was reduced by more than 50%. This mass loss happened because the bioplastics were composed from natural materials which easily digested by microbes. Bioplastics broke down into small pieces in 7th days, but the complete degradation occurred on the 9th day. After absorbing water from the soil, hydroxyl group in the cassava starch initiated the hydrolysis reaction; due to this reaction, cassava starch was decomposed into small pieces and quickly disappeared [36]. This also, reported by [35] bioplastic cassava starch-based with additional of glycerin and carbon has been successfully synthesized. XRD spectra clearly show an amorphous phase dominant was formed in bioplastics which were contributed from natural ingredients of the starch. The amorphous phase was decreased by increasing the amount of glycerin and carbon. Water absorption in bioplastics was strongly influenced by the amount of glycerin with the maximum absorption for 9 mL of glycerin in bioplastics. Degradation performance was increased with increasing the amount of carbon and glycerin with maximum values for 12 days put in the ground is 95.55% for the maximum amount of glycerin and carbon in bioplastics. For mechanical properties in the form of TS also shows the highest values for the highest amount of carbon and glycerin (9% glycerin and 1.5% carbon).

On the other hand, some researchers also reported about modification to cassava starch-bioplastic like, ozonated cassava [31], and addition oxidized starch [28] and nanofiber from cassava bagasse [29]. In [31] produced and evaluated films based on ozonated cassava starch. Ozonation resulted [17] enhanced mechanical properties, with increased TS, although decreased elongation and deformation during punctuation. This processes also increased the water vapour permeation and the oxygen permeation, also resulting in films with a more hydrophilic surface and less soluble. In general, the films presented good [22] sparency. Finally, the morphology of the ozonated films was more homogeneous, with increased relative crystallinity.

The research [28] has demonstrated that physic mechanical and thermal properties of bioplastic starch film can be enhanced with addition of oxidized starch. The opacity, moisture content and water solubility results show that material stored with these bioplastic films would be preserved better than that of the control. The mechanical properties indicated that the bioplastic films with oxidized starch have superior character as compared with the bioplastic without the oxidized starch. Similar with [29] all cassava starch films were transparent, flexible, and bubble free, potentially applicable for packaging, comparable to commercial films. TEM micrographs revealed that the nanoparticles had characteristic shape of nanofibril (diameter between 3 and 15 nm and

aspect ratio > 85). LCNF and Nanoclay were used to produce cassava starch films by solution casting with cassava starch, glycerol and water. Opacity and water absorption values of films reduced significantly and tensile stress of starch films with nano reinforcements were increased when compared to cassava starch. The water vapor permeability value was reduced for LCNF 0.65 and nanoclay 1.3, and a lower concentration of LCNF resulted in the lowest water vapor permeability value. The mechanical and barrier properties of starch films showed that LCNFs from cassava bagasse can be employed to reinforce starch films with potential uses in food packaging.

In a previous study, banana starch has been used for production of bioplastic sheets using glycerol as plasticizer by [25] Sultan et al. was reported the characteristics chemical bonding of the starch from the analysis of infrared spectra. The O–H stretching peak decreases from 3324 to 3317  $\text{cm}^{-1}$  upon the addition glycerol to low amylose starch which indicates a more stable hydrogen bond was formed between the glycerol and the starch molecules. The sharp peaks obtained at wavenumber 2850 to 3000  $\text{cm}^{-1}$  indicates the C–H bond stretching of  $\text{CH}_2$  groups in the starch structure. The peaks discovered at wavenumber 1580 to 1700  $\text{cm}^{-1}$  in both spectra assigned to the OH group deflection of water which was specifically due to hydroxyl groups bending mode in water molecules. The band at 1637  $\text{cm}^{-1}$  is a result of water adsorbed in the amorphous region of starch [25], and this result similar with study about starch and chitosan were used to prepare composites with PVA (polyvinyl alcohol) as an additive, glycerol as a plasticizer and glutaraldehyde as a cross linker. The composites were prepared by solution casting technique at optimum gelatinization temperature [38]. The broad band of St/Cs blends from 3500 to 3750  $\text{cm}^{-1}$  corresponds to the O–H stretching. The peak at 2900  $\text{cm}^{-1}$  corresponded to the C–H stretching, while the band at 1334  $\text{cm}^{-1}$  was the O–H stretching. The broad band at 2875  $\text{cm}^{-1}$  was the C–H stretching, while the band at 1541  $\text{cm}^{-1}$  was the N–H bending (amide II). A small peak at 1631  $\text{cm}^{-1}$  was due to the C=O stretching (amide I), and a peak at 990  $\text{cm}^{-1}$  shows the presence of an ether group in the film [25].

Based on [39] chitosan with banana starch (70:30) bioplastics containing 30% glycerol show low TS and high elongation just about  $\pm 3$  MPa and  $\pm 28\%$ . Compared with the control film, the nanocomposites reinforced with cellulose nanofibers present higher TS, Young's modulus, water-resistance, opacity, crystallinity, and thermal stability [41] The improved performance of the nanocomposites were attributed to, the chemical similarities between starch and cellulose, the fact of these components are from the same source, the nanometric size effect of the nanofibers, and the hydrogen bonding interactions between the fillers and the matrix. Regarding the mechanical treatment, as the

number of passages through the high-pressure homogenizer increased, the nanocomposites containing homogenized nanofibers displayed better properties than the nanocomposite produced with the non-homogenized nanofibers. On the other hand, the drastic mechanical treatment involving seven passages through the homogenizer led to defects in cellulose nanofibers, which act as crack initiators deteriorating the properties of nanocomposite. Furthermore, any agricultural residue, in this case banana peel, can be a source for processing cellulose nanofibers. The nanocomposites produced in this work could find applications in the food packaging industry [41].

The biodegradable plastic from sago starch is successfully developed and its reported by [44]. This biodegradable plastic is very environmental friendly based on FTIR result. In conclusion the sago starch based biodegradable plastic is degradable because FTIR test shows that after 14 days the major chemical inside the sago starch based biodegradable plastic is disappear. The mechanical compression test load versus elongation it reveal that the yield load is 60.2 N. That means sago starch based biodegradable plastic can applied load until 60.2 N. Lastly, the sago starch based biodegradable plastic have good acid resistance but poor alkalis resistance.

Research on the effect of chitosan filler levels and glycerol plasticizer on the properties and morphology of sago starch-based bioplastics has also been conducted by previous researchers, so [43] in this research will describe the manufacture processing of sago starch bioplastics using glycerol and sorbitol plasticizers with the addition of PVA and chitosan. The results obtained from this study are that glycerol plasticizers have better values in biodegradation testing, while sorbitol has better values in others. Glycerol plasticizer has the best value of 82.38% in the biodegradation test, while sorbitol with the best TS value is 16.12 MPa, 142.05% in elongation, tear strength is 12.729 kgf/mm, and water uptake value is 10.34%. This is related to the hydrophilic nature of glycerol and sorbitol, glycerol has a higher ability than sorbitol to absorb water so glycerol is more hydrophilic compared to sorbitol. It is also known that the combination of starch and PVA is a polymer that is hydrophilic because both are soluble in water so that if the plasticizer is mixed with starch-10 will increase the ability to expand in water. While [45] has demonstrated the potential of bioplastics composite based on starch/chitosan by additional Polypropylene. Bioplastic sample have a 10 rphous-crystalline structure from XRD analysis and the FT-IR spectrum shows that the functional groups of bioplastic has similarity with its constituent components. The maximum TS value of bioplastic at 68.41 MPa was obtained from the composition ratio 65/35. Biodegradation analysis shows that the synthesized bioplastic for composition ratio 65/35 degraded > 95% for 28 days.

In previous study [42] sago starch gel was sonicated for 0, 2.5, 5 or 10 min using an ultrasonic probe sonicator, then the non- and sonicated gel were treated using an ultrasonic bath. Ultrasonication duration for 10 min at 600 watts resulted in the best properties of sago starch film including highest thermal resistance, the lowest water vapor permeability, the highest transparency, and the lowest moisture absorption. The sonicated film showed a more compact and homogeneous structure, and had higher thermal resistance, and TS than the non sonicated film. Permeability to water vapor, opacity, and water absorption capacity of the films significantly decreased after ultrasonication. This present study provides further evidence that ultrasonication could become one simple, environmentally friendly process in the fabrication of sago starch film to improve its properties.

Application of rice straw, classified as an agricultural waste, for bioplastic production is reported [46]. Rice straw, a vegetable waste rich in cellulose, is used to produce a new biomaterial. The Naviglio extractor, already used in industrial applications, is employed for the first time in this work to pre-treat rice straw. After extraction, the solid matrix is treated only with trifluoroacetic acid (TFA), which is capable of co-solubilizing cellulose with other organic matter in rice straw. SEM analysis showed that the produced material is characterized by a uniform and compact matrix. After TFA evaporation, the obtained solvent cast film and sheet were continuous, flawless, flexible and resistant to tearing. Cellulose-based bioplastic possesses very good mechanical features, with TS and elongation at break over 43 MPa and nearly 6% for dried dumbbells and 11 MPa and 63% for wet samples, respectively. Based on the results, proposed bioplastic shows promising dual-shape memory effect. Although the recovery rates are between 60 and 80%, further studies are necessary to investigate and improve the production procedure of the bioplastic. Indeed, only a water-driven test was conducted for evaluating shape memory effect, and other external stimuli such as temperature change have not been explored. Regarding the mass loss test, the new bioplastic is demonstrated to be totally decomposed within 105 days after buried in soil. In terms of the sustainability of this material, data show that emissions and energies involved in its production are comparable to those of the thermoplastic starch and paper (cellulose based). These results highlight the possibility to obtain a new sustainable bioplastic. Rice straw can also be easily managed, because it does not require any separation from other waste. Therefore, application of rice straw for bioplastic production appears to be realistic.

All explanation shows that starches from different natural sources can be used, individually or combined, with and without the addition of different plasticizers, and kinds and amounts of natural fillers to produce a variety of different kinds of bioplastics boasting different physical and chemical characteristics. The differences in these properties will

allow the bioplastics to be suitable for varying applications. All the bioplastics produced were biodegradable and environmental-friendly, thus being a good substitute of plastics, and an efficacious way to alleviate the problem of plastic pollution [37].

However, the mechanical test results of bioplastic based on some starch are still very low and tend to be brittle, so it is necessary to add a composition such as natural fibers as reinforcement to increase the mechanical value of this bioplastic. Therefore, there are several starch-based bioplastics mixed with natural fibers which can be seen in Table 2.

There is a growing interest in materials science to use natural fibers (NFs) as reinforcement of polymer composites. Natural fibers have low density, good specific mechanical properties and readily available in many countries, recyclable, biodegradable and NFs use as reinforcement materials can help to reduce CO<sub>2</sub> emissions. Polymer composites with natural fibers have a low environmental impact, and depending on the polymer matrix, they can be labeled as eco-composites or green composite [54].

Among the reported studies, [55] were explored the main effects and interactions between treatment variables (temperature, alkali concentration) during mercerization on the final properties of those specific Pineapple Fiber (PALFs) were evaluated using a factorial design. Mechanical properties of single fibers were evaluated to assess if the fiber were a good option to obtain composites and physical chemical characterization was done to evaluate the changes in the structure provided by the mercerization process. In addition, the mechanisms observed for improvement of mechanical properties were studied. According to infrared analysis, the mercerization process induced significant variations in the lignocellulosic structure. After treatments, a higher crystallinity was observed caused by a more compact packing of the cellulose chains in the fiber. Mercerization treatments caused a reduction in the diameter of fibers. This reduction was attributed to two types of structural changes. The first structural change is related to separation of the elemental fibers when the middle lamella disintegrates. The second change is removal of non-cellulosic materials from the interfibrillar region promoting a greater interaction between the cellulose chains and finally produced a radial and longitudinal contraction in the fibers. Treatment increased the resistance of fibers from 288 (raw fibers) to 450 MPa (mercerized). The modulus of elasticity was modified for treated fibers and the treatment decreased stiffness of the fibers. Since mechanical properties of fibers were modified by mercerization, the fibers are expected to be useful as reinforcement in composites. Given their availability in some countries their use as reinforcement can add value to current wastes in pineapple harvest [55].

In previous study [56] potato starch-based nanocomposites reinforced with 1–4 wt% pineapple leaf (PALF)

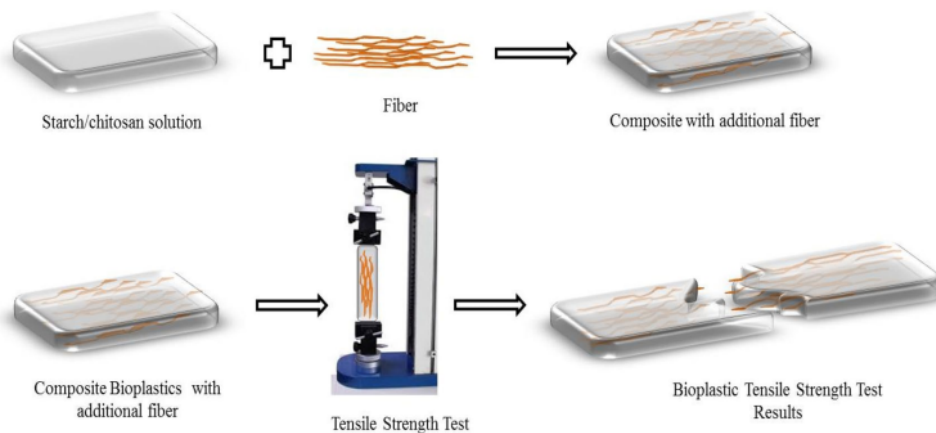
nanofibers were prepared by solution casting process. The morphological and structural analysis confirmed the formation of nanofibers from pineapple leaf. Dynamic mechanical analysis of prepared composites was analyzed thoroughly and from the inference conclude there is better interaction of Cellulose Nano Fiber (CNF) on to the thermoplastic starch matrix. Evaluation of constrained region shows that CNF had a tendency towards glycerol rich starch phase since the composite exhibit two relaxation phenomena as evident by tan delta curve. Modelling by Cole Cole plot shows an imperfect semicircle that further confirms the formation of heterogeneous system. Nanocomposites reinforced with 3 wt% of CNF shows better dispersion in all cases with low diffusivity and permittivity, high reinforcing efficiency, and high entanglement of nanofibers. In the study [57, 58] using pineapple leaf as fiber, the results were not much different from those shown in previous studies.

It can be seen that studies (Table 1 number 2) show a low TS properties with some glycerol mixtures. This is because the compound bonds in starch are very organic and brittle. So, in the study [57] pineapple microfiber (PLM) with chitosan was added and showed very large results, 51 MPa with 9% fiber and can decompose for 28 days. FTIR analysis shows the strong bonding formation was formed in bioplastics. Formation are C=C and C-H which was increases with increasing the amount of PLM in composites. The illustration can be seen in Fig. 2. It was investigate too by [64] this paper reports a new nanofibrillar cellulose (NFC) isolated from pineapple leaf fiber to be used as a filler in chitosan matrix to yield nanocomposite films. The research found that NFC concentration as much 3% from chitosan film resulted in optimum TS value. XRD analysis also showed the crystallinity increase from chitosan composite by adding 3% NFC into the chitosan matrix compared to control.

The increase of NFC above 3% caused a decrease of TS from chitosan film because aggregation has happened. This result was strengthened by the morphological analysis of film surface which was observed from SEM image. Water vapour transmission rate (WVTR) value decreased with the increase of the added NFC concentration. The increase of NFC in chitosan film has lowered the permeability of the composite film it cause water vapour route has increased to later diffuse into the film. Overall, at 54g NFC into chitosan film has successfully improved the mechanical and barrier properties of the composite film and is a good potential to be applied on biodegradable plastic for food packaging.

While in [58] the combination of short pineapple leaf fiber (PALF) and tapioca biopolymer (TBP) was capable of enhancing the mechanical properties of TBP biopolymer. Variance analysis revealed that 30% of the optimum fiber composition successfully increased the TS, tensile modulus  $T_m$ , flexural strength, and impact strength by 42%, 165%, 69%, and 10%, respectively. The PALF-TBP failure mode was comprised of a combination of broken fibers and no gaps between PALF and TBP, which showed better compatibility between PALF and TBP than PALF reinforced petroleum-based polymer composites. Overall, the incorporation of PALF as reinforcement in TBP has great potential as a renewable and biodegradable polymer.

A good number of fibers and synthetic matrices have been used making composites having a wide range of properties. As a matrix material, polypropylene (PP) has been extensively used with natural fibers in composite preparation [65]. PP with fibrous natural polymers of biomass origin is one of the most promising routes to create natural-synthetic polymer composites. So in previous study [59] fabricated PALF reinforced PP based composites with the variation of fiber percentage from 25 to 45% and different



**Fig. 2** Illustration of bioplastics with addition fibers before and after the tensile strength test

physico-mechanical properties were studied by conventional compression molding technique. TS, TM, Elongation at Break (Eb %), Bending Strength (BS), Bending Modulus (BM) and Impact Strength (IS) were evaluated. The 45 wt% PALF/PP composite exhibited an increase of 132% TS, 412% TM, 155% BS, 265% BM, and 140% IS with respect to the matrix material (PP). Fourier Transform Infrared (FTIR) Spectroscopy was employed for functional group analysis of PALF/PP composites. For all percentages of fiber, the composites demonstrated lower water uptake.

In fact, 30 vol% PALF composites have been recently investigated and found to present improved performance in terms of impact resistance and tensile properties [66]. Selected pineapple leaf fibers (PALF) significantly improve the strength of polyester matrix composites. This improvement corresponds basically to a linear increase up to 30 vol% of fiber incorporation and surpasses the flexural results with similar composites. The elastic modulus of the polyester composites is also increased with the volume fraction of PALF. SEM analysis indicates that PALF acts as an effective reinforcement for the brittle polyester matrix despite the weak fiber/matrix interface. In fact, the same fibers are well adhered to the polyester matrix but evidence of fiber pull out from the matrix indicates a relatively low interfacial shear stress. This is an important limitation for further composite improvement [60]. While, base on [61] show that the modified NFC increased surface hydrophobicity, and the contact angle of water droplet reached 114.49°. The surface modification improved interfacial compatibility and adhesion between NFC and polyester matrix, and made preparation of NFC (named as PF) uniformly dispersed in nanocomposite. As a result, the PF-based nanocomposite performed transparency, high tensile reinforcement (34.6% improvement at 1% PF), and good thermal resistance.

Santosh et al. were do the same thing like previous study with additional banana fiber. Banana and Pineapple leaf fibers have been used as reinforcement in polyester resin with various volume fractions. The results revealed that the thermal conductivity of composite decreased with increase in fiber content and the quite opposite trend was observed with respect to temperature. The specific heat capacity of the composite as measured by differential scanning calorimeter showed similar trend as that of the thermal conductivity [62].

To facilitate biodegradability in composites, polylactic acid (PLA) is often used as a matrix. PLA is one of the most extensively studied biopolymers mainly due to its high mechanical and thermal properties which are comparable to those of polystyrene and to its easy production from lactic acid monomer, which is derived from corn starch. PLA exhibits higher storage modulus and flexural properties, compared to polypropylene (PP) and is naturally decomposable to minimize environmental loads [63]. PLA-based

hybrid composites are prepared with coir fiber (CF) and PALF short fibers (30% vol.) as the reinforcing phase by the melt compounding and hot-pressing methods and their mechanical and thermal properties are investigated [63]. As a result hybrid composites demonstrated improved mechanical and thermal properties, due to the natural fiber reinforcement of the thermoplastic PLA matrix. Reinforcing the PLA matrix with CF and PALF improved the mechanical and thermal properties of the biocomposites. Hybrid composite (15% CF–115% PALF) out performed the other two hybrid composites as regards the tensile and flexural properties, as well as the storage and loss modulus. Moreover, its mechanical and thermal properties are comparable to those of plant/glass fiber reinforced polymer and conventional materials.

Sisal is the most commercially used natural fiber due to its better mechanical properties and chemical compositions. Similar to other, sisal fiber also has many advantages such as abundant, low cost, low density, recyclability, biodegradability, good strength and modulus. In addition these fiber, provide good acoustic and thermal insulation. Moreover, in comparison to other natural fiber, these fiber possess bright yellows color, higher strength, large fastener length, lower processing temperatures and so on. It is also noticeable that composites made by sisal fiber don't require more energy for their processing [67].

In previous study by [68] aimed to develop a facile preparation strategy of thermoplastic bio-based materials by combining continuous steam explosion (CSE) with periodate oxidation/borohydride reduction reactions. Sisal fibers were used as lignocellulose raw materials. The modification effects, mechanical properties, and thermoplastic of the products were investigated. In the CSE pretreatment, the aldehyde content of oxidized products was increased by a maximum of ~17%, and the yield of oxidized/reduced products was increased by a maximum of 25%. Furthermore, the TS of hot-pressed sheets was increased to 42.2 MPa, representing a 175% improvement compared with sheets without CSE pretreatment, and a decreased glass transition temperature of ~132 °C.

Results indicated that CSE pretreatment was favorable for improving the modification efficiency and uniformity of sisal fibers, simultaneously achieving much better mechanical properties and thermoplastic. Appropriate oxidation level of sisal fibers also achieved the better performances. The combined strategy was facile and scalable, and might significantly impact the widespread preparation and utilization of thermoplastic bio-based materials [68].

The advantages of natural fiber over traditional reinforcing materials such as glass fiber is acceptable specific strength properties, low cost, low density, high toughness, good thermal properties and biodegradability. The combination of sisal fiber and glass fiber for reinforcing epoxy polymers gives enhanced mechanical properties. The work deals

with the utilization of sisal fiber and glass fiber reinforcement in polymer. The objective of the work is to compute mechanical properties such as tensile and hardness of randomly oriented sisal fiber and glass fiber reinforced epoxy composites. In this work, sisal fiber % weight is varied from 2 to 8 with constant 2% weight of glass fiber. From the work, it was found that ductility, modulus of elasticity and hardness gradually improved [69].

In earlier studies researchers have reported that the sisal fiber increases the heat capacities of the thermosets (epoxy) than the glass and other manmade fiber. Polypropylene is thermoplastic material looks white in appearance usually available in pellets. Polypropylene is a versatile material. Its strength varies from 15 to 35 MPa. Polypropylene is mixed with polyethylene to improve its moulding properties which is referred as moulding grade polypropylene. The density of the PP is around  $0.91 \text{ g cm}^{-3}$ . It is hydrophobic material having excellent resistant to most of the chemicals. Its melting point is also high and hence usually used as insulator in electrical devices [70, 71]. So, [72] was investigated the effect of reinforcing sisal fibers on the mechanical and thermal properties of polypropylene composites. Nano Fiber Cellulose (NFCs) were developed in powder form and compared with short fiber reinforced composites. NFCs composites have better heat absorption and rejection capacities than the pure polymers. However, these composites showed lesser mechanical strength when compared to the pure polymer. The thermal analysis through differential calorimetric analysis (DSC) shows that there are no much appreciable changes in the endothermic peak and the exothermic peak irrespective of fiber loading. The powdered form and 10 mm fiber composites have showed better heat capacities and mechanical strengths when compared to the 20 mm fiber. The thermal results indicated that, heat absorption capacity of the composite is maximum for 10 mm and 10% of fiber. The untreated fiber composites showed lesser strength and the reduction in heat capacities is up to 33%. It indicates that the elimination of water resulted in lesser heat capacities. It can be concluded that as the weight fraction of the fiber increases in the matrix the strength of the composite decreases in a particular length of the fiber. The 20 mm fiber showed poor mechanical strength and lesser heat capacities. Though presence of sisal fiber increases the heat capacities of the polymer but mechanical strength should be compromised. The microstructural analysis results showed that polypropylene matrix with powder form and short sisal fiber of 10 mm improved the adhesion strength and TS of the composite [72].

Similar with [72], Munde et.al was presents a systematic approach to evaluate and study the effect of fiber loading on the mechanical (tensile, flexural, impact and hardness) and morphological (SEM) properties of short sisal fiber reinforced polypropylene polymer-based composites. Short sisal

fibers of 3 mm length are incorporated into Polypropylene (PP) in three different fiber loading condition (10, 20 and 30). Composites are fabricated through extrusion followed by injection moulding process. Results are compared with neat PP. Mechanical characterizations are done as per ASTM recommendation and average of five is reported. Experiments show that, at higher fiber loading condition (30%), enhanced mechanical properties are recorded. Fractography is done of the tested specimen through electron microscopy. Images reveal that, fiber entanglement enriches the mechanical interlocking and polymer anchoring inside the composite system. In addition, the elastic behaviors of the produced composites are studied using Representative Volume Element (RVE) method. A good dealing between the models developed with the experimental values is observed [73]. In the study [74] using PLA as a matrix, the result were not much different from those shown in previous studies. Study presents the recyclability analysis of PLA/Sisal biocomposites comprising sodium bicarbonate treated Sisal fibers (30 wt%). The biocomposites were recycled (8 times) using the extrusion process. The TS of injection molded biocomposites declined by 20.9% up to the third recycle. The dynamic mechanical analysis revealed a severe reduction in storage and loss modulus beyond third recycle. The morphological and thermal characterization of recycled biocomposites also revealed severe fiber and matrix degradation. The recycling of PLA/Sisal biocomposites beyond third recycle is not recommended. However, the biocomposites recycled up to third recycle can be used for making products for low to medium strength non-structural applications.

Bamboo fiber reinforcement composite materials have been used as reinforcement in composite processes. Different researchers concluded that the mechanical properties of bamboo reinforced composite are affected by different parameters. TS, tensile modulus, impact strength, compression strength, and moisture content reduction of composites are significantly affected by filler particles, bonding agents, processing techniques, size, and structure of fibers. Temperature, fibers orientation, volume and weight ratio or composition of fiber or additive materials are other parameters that affect composites. In generally, properly controlling and balancing the parameters is a better methodology to obtain excellent mechanical properties of bamboo reinforced composite materials [79].

In previous study, Yusof et.al, investigate the mechanical properties and biodegradability of green composite by applying chemical treatments on the bamboo fibers that act as reinforcement on the tapioca starch for making possible this biocomposite to compete with the petroleum-based plastics in the market. Different type of chemical treatments leading to increase the tensile and flexural strength of composite. Alkali treated starch/bamboo fiber reinforced composites has been proved to have

better mechanical among the samples. Throughout all the experiments, the characteristics of the composites show that the chemical treatment will have significant effect on the mechanical properties of the composites [75].

Similar with previous study, [76] shown biocomposite films was prepared using bamboo fiber to reinforce modified and unmodified red seaweed (*SW Kappaphycus alvarezii*), resulting in improved mechanical characteristics of 109.1 MPa TS, 55.4 MPa Young's Modulus and 22.3% stretch ability prior to breakage at the optimum value of 15% bamboo fibers loadings for unmodified biocomposite. There was general improvement in the fiber/matrix interface of the modified SW based composite films over the biopolymer films from unmodified SW bamboo reinforced films resulting in improved water vapour barrier as the fiber load increases. FTIR analysis validates the effective interaction of both the bamboo fibers and the seaweed matrix without any significant biochemical alteration of the SW within the frameworks of composite films. SEM characterization indicate that heterogeneous surface modification of the biopolymer film increases with the fiber loading are represented in Fig. 3.

Among the natural fibers available, bamboo fiber has relatively high strength. Polylactic acid (PLA), one of the well-known biopolymers, has been used as a matrix in order to produce totally biodegradable biocomposites. In previous study, bamboo fibers were compounded with PLA by a twin screw extruder. The bamboo fiber reinforced PLA composites were then manufactured via the compression moulding method. The influences of screw speed and die temperature during extrusion on the mechanical properties, the tensile and flexural of the biocomposites, were studied. The effects of fiber content and fiber length were also investigated. Taguchi experimental design approach was adopted to determine the optimum set of conditions to achieve the "best" mechanical properties of the composites. Tensile and flexural properties were characterised based on the D638-10 and D790-10 standards, respectively. It was observed that the fiber aspect ratio and fiber content significantly affected the mechanical performance of bamboo fibers reinforced PLA composites [77]. In the study [78] using a bamboo as fiber, the results were not much different from those in previous study.

Sugarcane bagasse fiber is a kind of fiber that has not been used much. This fiber has a high cellulose content and better as a filler in composite. It is seen as a good reinforcement in the starch-based matrix. This can be explained by the hydrogen bonds formed between the two materials. Sugarcane bagasse fiber is classified into high cellulose content which is around 85% so it can produce a lot of -OH bonding. The -OH bonding will interact with -OH from starch and produce good mechanical properties [80].

In a previous study, tapioca starch adds with sugarcane bagasse fiber (SBF). The addition of SBF into the tapioca matrix serves as a structural reinforcement of composites bioplastics [80]. The fabrication of composite using the solution casting method. The variation time duration of ultrasonication on composite bioplastic (tapioca starch and 1 wt% bagasse fiber) includes 0, 5, 10 and 15 min. The maximum TS value is in the variation 15 min ultrasonication (2.5 MPa) and this result is higher than the sample without ultrasonication for 1.1 MPa. This makes the composite bioplastics more brittle. The change in morphological structure is proof that ultrasonication is effective in improving fiber dispersion in the matrix and increasing adhesion bonding between the fiber and the matrix. The results of moisture absorption rate also prove that the addition of ultrasonication makes composite bioplastics become more resistant to water vapor and moisture. However, this study still have a low TS value than as can be seen at Tabel 1 number 3. This may due the lack of fiber concentration given. So, in the next work, increasing the concentration of fiber using a similar starch can increase its mechanical value.

There are some studies reporting the influence of modified fibers in the reinforcement of starch-based composites. Starch acetylation is the most common chemical modification by which a part of the hydroxyl groups of an hydroglucose units is substituted by acetyl groups, altering the starch molecular structure. Several studies have found that starch acetylation can improve the mechanical barrier properties of biocomposites [83]. Nonetheless, in order to improve the reinforcement of acetylated starch-based biocomposites, natural fibers may incorporated. Therefore, [81] on this work is to evaluate the effect of acetylated fiber and glycerol contents on the mechanical, physical and microstructural properties of biocomposites composed of acetylated corn starch and acetylated sugarcane fiber. Due to acetylation and possible interaction between matrix-fiber, there was an improvement in water resistance; while the mechanical properties were enhanced by increasing acetylated corn starch with acetylated sugarcane fiber (FC) up to 12.0%. Biodegradability recorded a range of 24.2–39.3%. Microstructural analysis evidenced the extrusion process effect, chemical modification and new interactions formation. It was found that an optimum blend was of FC = 12.0% and GC = 24.0%. The acetylation of both sugarcane fiber and corn starch allowed us to obtain eco-friendly materials with good mechanical properties and water resistance [81].

Mulinari et al. were investigate too about sugarcane bagasse fiber with polypropylene composite [82]. It was used maleic anhydride grafted polypropylene (PPg-MA) as coupling agent. Fibers were characterized by the techniques of scanning electron microscopy (SEM) and Fourier Transform Infrared (FTIR) spectroscopy. The chemical composition of fibers was also evaluated. PP was mixed to fibers, in

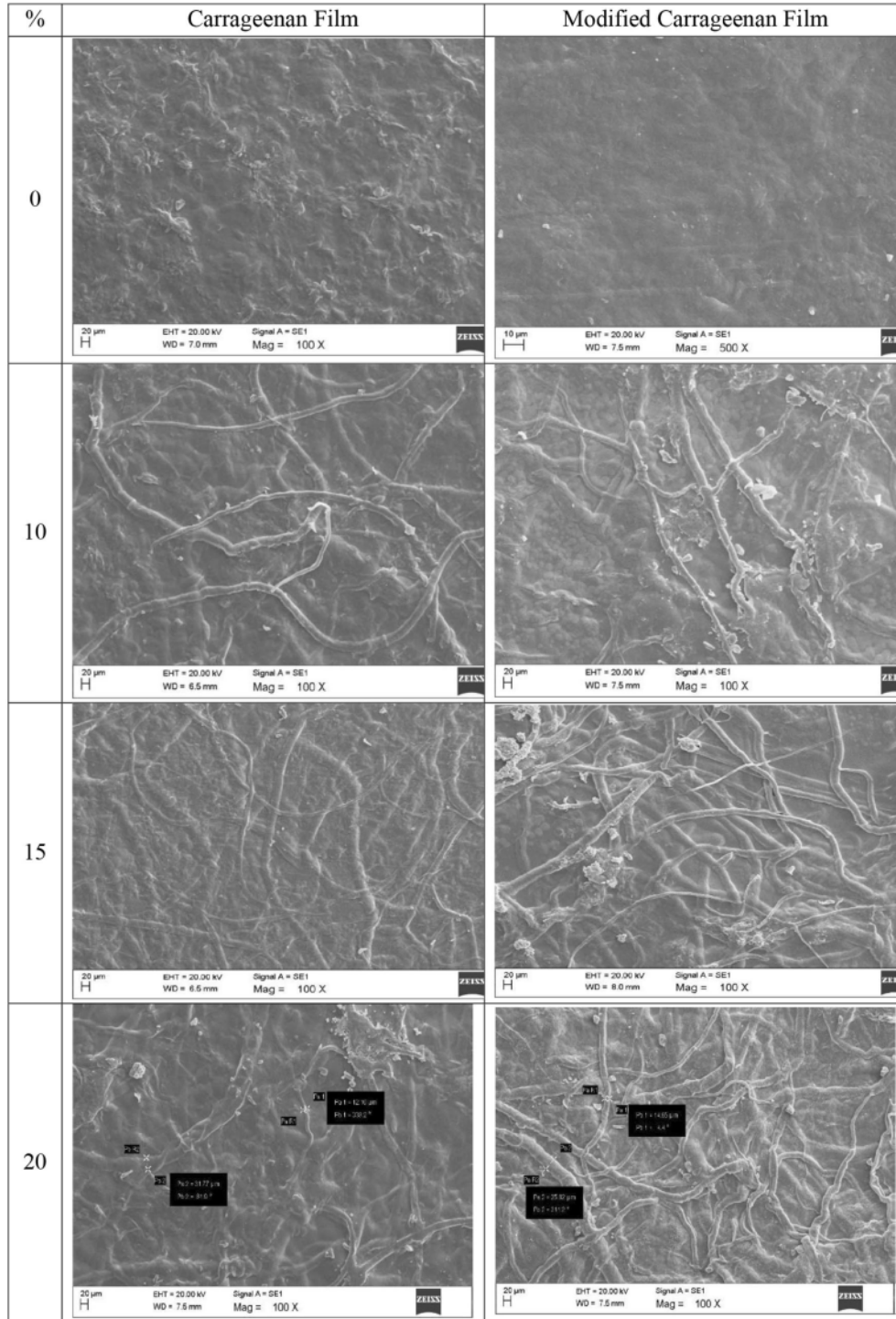


Fig. 3 SEM micrograph of surface morphology with varying bamboo pulp content at 100× magnification [76]

proportions of 5–10% (wt/wt) using a thermokinetic mixer model MH-50H. The pre-treatment influenced the mechanical properties of composites: the tensile modulus of elasticity of PP increased 66.1% in the composite with 10 wt%, with only 29.8% when PP was reinforced with the same amount of untreated fibers. On the other hand, the use of PP-g-MA influenced directly the mechanical properties of composites: the tensile modulus of elasticity of PP increased 77.1% in the composite with 10 wt%. Tensile tests results evidence good modulus for the composites with pre-treated and untreated fibers, similar to that of the reference containing P-g-MA as compatibilizer.

Agricultural wastes, including cocoa pod husk (waste from the chocolate industry) and sugarcane bagasse (waste from the sugar industry), are increasing day by day. The development of food packaging biofilms from these two wastes could be beneficial to the environment and human. Therefore, [84] was conducted to develop biodegradable plastic films by using cocoa pod husk and sugarcane bagasse. Cellulose and fiber were extracted from cocoa pod husk and sugarcane bagasse, respectively. The physicochemical properties for all bioplastic concentration ratios were determined in terms of sensory evaluation, drying time, moisture content, water absorption and water vapor permeability. From the observation and analysis of the physicochemical properties of bioplastic, found that the most suitable bioplastic film for food packaging goes to the combination of 75% cellulose and 25% fiber bioplastic, as it demonstrated the lowest water absorption percentage and water vapor permeability. In the study [85] The cellulose microfibrils (CMF) from water hyacinth (WH) fiber as a filler in sago starch (SS) bio-composites was investigated. The CMF was isolated by pulping, bleaching and acid hydrolysis methods. The addition of CMF in sago matrix was varied i.e. 0, 5, 10, 15 and 20 wt%. Bio-composites were made by using solution casting and glycerol as a plasticizer. The addition of CMF-WH sago starch biocomposites lead to the moisture barrier, crystallinity, and thermal stability increased; it is due to the pure sago starch film was more rapidly degraded than its biocomposites. [16]

While in [86] Biocomposites with a corn starch-based biodegradable polymer as matrix and 10 wt% vegetable curauá fiber were processed by injection molding and were submitted to reprocessing up to ten cycles with or without 3 wt% of maleic anhydride grafted polypropylene as coupling agent. Curauá fibers have increased hardness, impact and TSs as well as increased tensile modulus and decreased elongation at break of the biocomposites with respect to starch-based matrix and these properties slightly decreased or no considerable changes were observed with the reprocessing cycle increase.

From some descriptions of the studies above, it can be concluded that the addition of fibers to the bioplastic

composition will increase the TS, elasticity of the bioplastic itself and facilitate the degradation process because it easily absorbs water but over time, researchers have also carried out several experiments, by adding nanoparticles to the bioplastic composition to further add the mechanical value of the bioplastic and to inhibit a bit of the biodegradation process, which has applications for food packaging. Several studies using nanoparticles in bioplastic compositions can be seen in Table 3.

In previous study, jeneng starch bioplastic were made with glycerol as the plasticizer [4] ZnO with varying concentrations of 1, 3 and 5% were added into the biopolymer to improve the mechanical properties. The highest TS value was obtained at the concentration of 3% ZnO starch with the addition of 3 ml glycerol, which was 2.74 MPa. Percent elongation increases with increasing concentration of plasticizer, at a concentration of 1% ZnO with the addition of 6 ml glycerol, which is equal to 4.5%. The lowest water absorption is obtained at 1% ZnO concentration with the addition of 1 ml glycerol of 2.9. The process of plastic film degradation occurs for 30–39 days. The results of the DSC thermogram shown the thermal degradation temperature of plastic with 6% ZnO has the highest value of 390.56 °C far above the addition of 1% and 3% ZnO with values, 361.53 °C and 362.15 °C. This shows that plastics with higher ZnO concentrations have better thermal resistance. At the same nanoparticles, [18] study a new nanocomposite film and coating based on chitosan-carboxymethyl cellulose-oleic acid (CMC-CH-OL) incorporated with different concentrations (0.5, 1 and 2%) of zinc oxide nano particles (ZnO NPs) have been suggested as a packaging material to increase the shelf life (microbial and staling) of sliced wheat bread. Control exhibited the highest firmness over 15 days of storage in all other samples. The results of microbial tests revealed an increase in microbial shelf life of sliced wheat bread from 3 to 35 days for CMC-CH-OL-ZnO NPs 2% in compared to the control. All active coatings lessened the number of yeasts and molds in sliced bread during 15 days, and further improvement in antimicrobial properties obtained for coatings contains 1–2% ZnO NPs with no fungal growth during 15 days.

Abdullah et al. were investigate the reinforcing effect of ZnO on the physical, mechanical and antibacterial properties of starch-based bioplastic. Bioplastic was prepared by melt-mixing starch and glycerol (3:1, w/w) with ZnO (1%, 2%, 3%, 4% and 5%, w/w). Bioplastic density and water contact angle increased with the increase of ZnO concentration. Bioplastic with the addition of 4% ZnO showed the lowest moisture content of 3.45%. The decomposition temperature of bioplastic with ZnO increased slightly which indicated the higher stability. Mechanical properties evaluation showed that bioplastic with addition of ZnO had higher TS than that without ZnO where 4% ZnO exhibited the highest

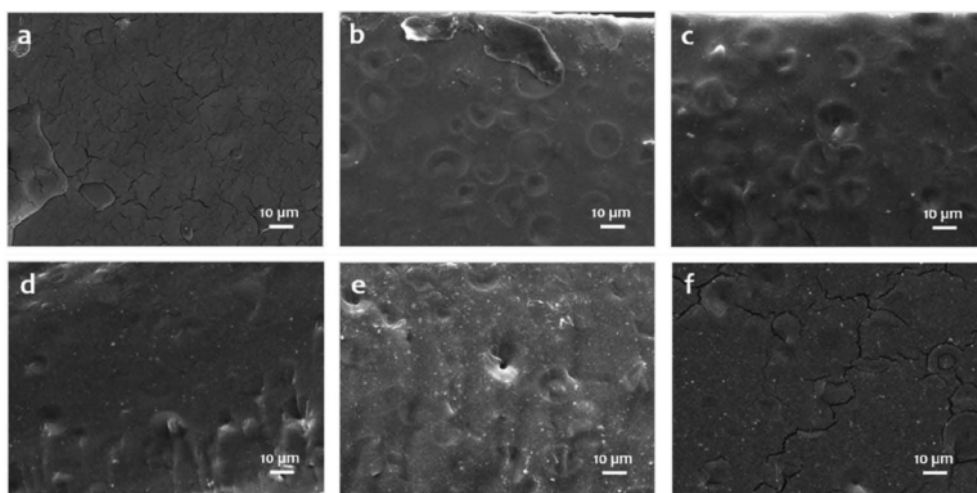
TS of 10.29 MPa with elongation of 5.69%. In addition, biodegradability tests of bioplastic showed that the growth of microbes decreased in the presence of ZnO due to the nature of ZnO as an antibacterial compound. The results showed that ZnO played a key role in reinforcing the physical, mechanical and antibacterial properties of starch-based bioplastic [33]. The addition of ZnO to starch-based bioplastic can be seen by SEM micrographs in Fig. 4.

Similar with previous study, [46] was investigate the influence of zinc oxide nanofiller on the mechanical properties of bioplastic cassava starch films with variations of zinc oxide and glycerol in biopolymer film by casting technique. The result showed that the TS was improved significantly with the additional of zinc oxide but the elongation at break (EB %) of the composites was decreased. The maximum TS obtained was 218 MPa on the additional of zinc oxide by 0.6% and plastilizer by 25%. Based on data of FTIR, the produced film plastic didn't change the group function and it can be concluded that the interaction in film plastic produced was only a physical interaction. Biodegradable plastic film based on cassava starch-zinc oxide and plasticizer glycerol showed that interesting mechanical properties being transparent, clear, homogeneous, flexible, and easily handled.

In a previous study, chitosan-banana starch bioplastics were made with glycerol as the plasticizer [39] ZnO with varying concentrations of 1, 3, and 5% were added into the biopolymer mixtures before casting in order to improve their mechanical properties. It turned out that 3% ZnO significantly increased the bioplastics TS up to ~36 MPa. Also elongation and swelling percentage were decreased as ZnO concentrations increased. Bioplastics prepared with 3% ZnO were degraded within relatively short time in 90 min. These


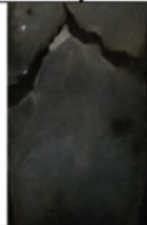
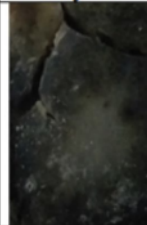





chitosan-banana starch bioplastics reinforced by ZnO seems quite promising to substitute some commercial conventional plastics. At the same nanoparticle [33, 37] This research aims to study the effects of additive (glycerol and zinc oxide) addition in the characteris [33] of antimicrobial activity and biodegradability bioplastic from chitosan and Kepok banana peel starch. Bioplastics were characterized their antimicrobial activity using agar diffusion method (zone inhibition assay) and biodegradability test using microbe (EM4). The result showed the optimum composition of bioplastic is chito: 47.4–30% starch—5 mL glycerol—5% ZnO gives the good antimicrobial activity towards gram positive and gram negative bacteria, and this bioplastic will be degraded within an h: 25 and 12 min. Thus, this bioplastics may have potential to be use for food packaging by having biodegradable properties and also inhibit bacterial growth.

In this way, composite materials containing nanoparticles inorganics can generate innovative high-performance materials. The inorganic nanoparticles keep exceptional interfacial interactions in bioplastics, and significantly improve their properties. According to literature, all mechanical and thermal properties of bioplastic composites improve with the addition of nanoparticles inorganics [7]. The starch-based with the addition of 0.5% of silica powder bioplastics were produced by casting and characterized by color analyses, transparency, opacity apparent, humidity, thickness, TS, elongation at break, FTIR, DSC, SEM, and biodegradation assay. The addition silica powder improved the elongation at break of the corn starch-based bioplastics. The sample presented increased from 59.2% (without silica powder) to 78.9% (with silica powder). For potato starch bioplastic the addition of 0.5% of silica powder didn't improve elongation



**Fig. 4** SEM micrographs of bioplastics with **a** 0%, **b** 1%, **c** 2%, **d** 3%, **e** 4% and **f** 5% ZnO content [33]

**Fig. 5** Degradation of starch bioplastic and composite bioplastic [20]

	Day 1	Day 7	Day 15	Day 30
Starch Bioplastic				
Weight loss	0%	13%	54%	81%
TiO <sub>2</sub> Composite Bioplastic				
Weight loss	0%	7%	32%	64%

at break but increased the thermal resistance. Increased until 17 °C until 11 °C. The bioplastics of potato starch were biodegraded in 5 days, and those of corn starch took almost 40 days. Silica powder inhibited the growth of fungi in starch bioplastics [88].

Zhang et al. [7] demonstrated that the size of the silica nanoparticles played an important role in the physical performance and mechanical properties of potato starch films. The 100 nm silica nanoparticles were uniformly dispersed and there was an improvement in the mechanical properties of the potato starch bioplastics. The antibacterial experiment [25] indicated that the nano-SiO<sub>2</sub>/potato starch films exhibited good antibacterial activity against the *E. coli* and was less efficient against the *S. aureus*. The effects of the preservation properties of the nano-SiO<sub>2</sub>/potato starch films on the white mushrooms were remarkable; and those of the 100 nm films were superior.

In the study by [20] it can be ascertained that starch-based bioplastics and composite bioplastic have been successfully characterized by various analysis. The composite bioplastic is stronger than starch bioplastic with increased TS and reduced elongation. From FTIR analysis the absorption band of the C–H and C–O–H at 2932 cm<sup>-1</sup> and 1152.92 cm<sup>-1</sup> for the starch bioplastic shifted to lower wavenumbers at 2926.31 cm<sup>-1</sup> and 1151.54 cm<sup>-1</sup>, which is confirmed the electrostatic interaction between the starch chains and titanium dioxide. By adding TiO<sub>2</sub> to starch bioplastics, its properties have been improved. The preparation of starch bioplastic and composites bioplastic with better thermal, mechanical and chemical properties is a significant achievement. These products can be a appropriate alternative for the existing conventional plastics for its high biodegradable

properties with suitable thermal and mechanical properties. Also starch is a renewable resource, cheap and easy to modify. Titanium dioxide nanoparticles have antimicrobial properties and composite bioplastics can be considered suitable for the food and pharmaceutical industry considering the experimental results as can be seen in Fig. 5.

After seeing the results of research by adding ZnO or TiO<sub>2</sub> nanoparticles, the mechanical properties of each bioplastic improved. In this study [21] aims to make an eco-friendly plastic and to improve the quality of bioplastic by adding Zinc Oxide and Titanium Oxide nano particles, the influence on the mechanical properties, surface morphology, and the decomposition rate of bioplastic was investigated. Based on the results, it can be concluded that the addition of ZnO and TiO<sub>2</sub> nanoparticles can increase the mechanical strength and elongation of bioplastics, but on the contrary rate of bioplastic degradability decreases with the addition of ZnO and TiO<sub>2</sub>. The highest TS 20 MPa was obtained of percentage composition of nano ZnO and nano TiO<sub>2</sub> 25:75. On the other hand, the highest elongation (29.3%) was obtained of percentage composition of nano ZnO and nano TiO<sub>2</sub> 0:100 and biodegradability process during the 3 days showed that the easier bioplastic film to decompose in the soil are bioplastic films without the addition of a substance of nano ZnO and nano TiO<sub>2</sub>.

In this research [22] bio films were prepared by casting method as a replacement for the traditional petroleum based plastic. It widened the use of potato starch based film blended with CaCO<sub>3</sub> (PSBBCCF) in food packaging film as expected. PSBBCCF was gelatinized in the presence of water and varying amount of glycerol. Potato starch blended with 50% (w/w) of calcium carbonate mechanically

to enhance mechanical and physical properties. PSBBCCF were dried under controlled conditions. Physicochemical properties such as water absorption capacity, hardness, solubility in water and mechanical properties of the PSBBCCF were investigated. Results revealed that by increasing plasticizer the crystalline of starch in the film were decreases. PSBBCCF maybe use an alternative material for synthetic plastic to avoid waste generation and disposal. At the same nanoparticles [30] the addition of precipitated calcium carbonate (PCC) into a cassava starch matrix increased the TS of the resulting bioplastic/PCC composite with an optimum value with the addition of 4% (w/w) of PCC. This resulted in a composite with TS and modulus of elasticity of 3.38 MPa and 465.15 MPa, respectively. Analysis of TGA graph shows that the addition of PCC improved the thermal stability of the composite. The moisture absorption test indicated that the composites were hydrophilic, although the water absorption properties were reduced from 70.59% to 53.33% after the addition of 10% PCC. The XRD results test showed bonding between the tapioca starch cellulose and PCC. After PCC content exceeded 4% distribution was no longer even and aggregation occurred decreasing the TS.

## Conclusion

From the table classifications above, it can be concluded that bioplastics are one of the most innovative materials that are bio-based and biodegradable. As explained, if starch-based bioplastics still have weak mechanical properties, it is necessary to provide fiber as reinforcement and several studies have shown that with the presence of nanoparticles, the resulting bioplastic mechanical properties are much better. So, it is hoped that further research will be conducted, in order to combine these three materials to obtain a strong but environmentally friendly biocomposite result.

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## Declarations

**Conflict of interest** The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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