

Review on the characterization of starch biocomposite films in packaging application

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Abstract

As an alternative to advocating biodegradable packaging to reduce plastic wastes, starch-based materials are proposed to be used as an element to be integrated into the packaging due to their abundance in nature. Starch-based materials can be converted into any desired shape and size, therefore, making it the perfect basis for eco-friendly packaging. The availability of starch readily in nature from numerous resources including cassava, maize, and potato enables it to be a low-cost raw material. However, starch-based packaging is often deemed weak and brittle thus, it requires reinforcement. It can be from various biological origins including cellulose, fibres, and nano clay producing starch biocomposite materials. Starch biocomposites are frequently studied to be employed in packaging due to their biodegradability which when used in the packaging can be directly degraded into the environment. The form of films for these biocomposites is often analyzed and these biocomposites films possess improved mechanical and optical properties such as higher tensile strength and lower density which resemble the conventional petroleum-based plastics. Studies have included the outcomes of fabrication and characterization analyses of biocomposite film based on the morphological, optical, thermal, barrier, and mechanical properties. Therefore, this paper aimed to review the characterization of the starch-based biocomposite films that can be applied in packaging based on the analyses done by previous researchers.

1. Introduction

In 2019, the worldwide production of petroleum-based plastics reached 370 million tons, with packaging accounting for 39.6% of that total (Plastics Europe, 2020). Since the usage period of 60 per cent of plastic products and components is between 1 and 50 years before they finally occupy the landfill, the quantity of produced and used plastics is gradually outmatched by the number of gathered plastics every year (Europe, 2020). Hence, environmentally friendly products have been promoted as a solution to the mountains of plastic wastes accumulating in our landfills and the main characteristic of the products is biodegradable.

Biodegradability is the ability of the product to be degraded with organic wastes into the soil without increasing the pollutant and occurs in the presence of microorganisms (Vinod *et al.*, 2020). Therefore, when biodegradable products are used and disposed of, they would not occupy the landfills for a long time owning it

to their biodegradability which is opposite to petroleum-based plastics as they would require many years to be degraded (Roy *et al.*, 2015). Thus, notable attention must be given to packaging when purchasing an environmentally friendly product. Hence, the concept of biological-based materials for packaging has been established as an alternative to petroleum-based plastics which are not degradable.

The abundance of starch in nature enables it to become the main resources to be utilized in designing environmentally friendly packaging. They are relatively cheap, easily worked, and accessible qualifying them to be a versatile raw material for production (Carvalho, 2008). There are various sources of starch such as cassava, maize, and potato. They are carbohydrates with various ratios of amylose and amylopectin in which the linear structure of amylose enables it to mimic the structure of the conventional synthetic polymers while the amylopectin acts as the core crystalline element (Zou

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et al., 2012). The disruption and plasticization of amylose and amylopectin in starch known as gelatinization is done to acquire thermoplastic starch in the presence of water and glycerol as plasticizers (Cyras *et al.*, 2008). Thermoplastic materials made of starch have been proposed as an alternative to non-biodegradable petroleum-based plastics (Li *et al.*, 2015). However, the hydrophilicity properties, melt instabilities, and crystallization behaviour of most starch-based products have become the reasons for them to be deemed weak and brittle thus requiring reinforcement (Fatin *et al.*, 2018). Hence, the development of starch as biocomposite films is a good approach to be used for packaging.

Biocomposites are materials that are compounded with one or more phase(s) which are derived from green materials (Noor Zulaika *et al.*, 2020). The biocomposites are often reinforced with other biological materials such as fibres and its matrices are usually polymers from abundant resources such as starch (Fowler *et al.*, 2007). They are mostly employed in various fields including wood-plastic composite (WPC) for constructions, kenaf-PLA biocomposite mats for automotive interiors, and sports equipment including canoes and snowboards from flax biocomposite owing to their improved tensile strength, lightweight and environmentally friendly materials (Quarshie and Carruthers, 2014). Hence, there is various research that studies the potential of biocomposite films to be used in packaging.

In designing packaging, there must be specifications and standards that can enable the materials to be incorporated into the design. Thus, characterization analyses for biocomposite films were to be done to quantify the results and therefore, enable the materials to be used as packaging. The films must possess an acceptable mechanical property, and applicable permeability to water, vapour, and gases in which they are conditional to the product characteristics and proposed end-use application (Alves *et al.*, 2006). Therefore, this paper aimed to review the characterization of the starch-based biocomposite films that can be applied in packaging based on the analyses done by previous researchers.

2. Fabrication of film

Fabrication of biocomposite films as stated in Table 1 can occur through various processing which differ according to the type of reinforcement, plasticizers, polymers, and the means of applications (Vinod *et al.*, 2020). In compression moulding, gelatinization of starch occurs first, then followed by an expansion of the starch mixture in a mould by compression at a designated temperature and pressure for a period of time, and

finally, it is dried to produce soft and stretchable films with an improved optical property (Mangaraj *et al.*, 2019). Comparably, for the solution and solvent casting methods, the starch and plasticizer are mixed to form gelatinization, which is then heated and cooled before being poured onto an acrylic plate and allowed to dry until a constant weight is attained. The only difference between solution and solvent casting methods is that, in the solvent casting method, chloroform is used as a solvent to dissolve the starch which is the PLA before gelatinization (Anuar *et al.*, 2017). Conversely, for the film casting method, a film casting machine is utilized where the solution is poured into the machine plate with assigned speed, temperature, and blade distances to produce films with the desired thickness (Mustapha *et al.*, 2019). Similarly, for the extrusion method, a starch biocomposite mixture is extruded with a slit die in a machine to develop the films for the extrusion method (Yu *et al.*, 2013).

Among all the processes, the extrusion method can bring multi-level changes to the structure of the starch as well as it can degrade the amylopectin molecules which can further disrupt the crystalline and granular structure of starch (Li *et al.*, 2015). However, the mechanical properties and the surfaces of the biomaterials produced by the extrusion process are also affected by the speed of extrusion as too much speed can ruin the structure (Suryanto *et al.*, 2020).

For nonsolvent induced phase separation (NIPS), the fabrication process includes the biocomposite mixture of starch and its reinforcement to be cast on an impermeable support sheet as a thin film before drying (Asgarpour Khansary *et al.*, 2018). Similarly, for hydraulic pressing, the starch biocomposite mixture is mixed in an external mixer prior to casting by using a hydraulic presser at a designated temperature and pressure (Irwanto *et al.*, 2020). On the other hand, in milling processing, the starch mixture is cast in a two-roll milling machine with a lift-to-drag ratio of 1:40 in a high-temperature condition (Roy *et al.*, 2015).

Glycerol as a biodegradable plasticizer is typically utilized in most fabrications as it can lower the intermolecular strength of the molecules, increase the polymer chains' mobility, lower the temperature of glass transitions, and amplify the permeability (Nur Humairah and Zuraida, 2012). The incorporation of plasticizers helps in improving the mechanical, chemical, and thermal properties as plastination of starch are enabled by the hydrogen bonding between the starch and plasticizers (Carvalho, 2008; Vinod *et al.*, 2020). Mochamad *et al.* (2020) deduct that the ultrasonication process helps the breakage of hydrogen in the cellulose

Table 1. Fabrication process

Type of starch	Reinforcement/ Fillers	Plasticizers or crosslinking agents	Methods	References
Polylactic acid (PLA)	Banana stem microcrystalline cellulose (BSMCC)			Suchaiya and Aht-Ong (2011)
Maize	Water		Compression molding	Li <i>et al.</i> (2015)
Thermoplastic corn	Bacterial cellulose	Glycerol		Santos and Spinacé (2021)
Rye flour	Cellulose	Citric acids		Beigmohammadi <i>et al.</i> (2020)
Potato	Polyvinyl alcohol (PVA)	Yerba mate	Electrospinning	López-córdoba <i>et al.</i> (2019)
Cassava	Acerola residues		Extrusion and injection molding	Reinaldo <i>et al.</i> (2021)
Potato	Chestnut husks		Extrusion molding	Torres <i>et al.</i> (2019)
Cassava	Carboxymethylcellulose (CMC)	Glycerol		Fatin <i>et al.</i> (2018)
Sago	Montmorillonite (MMT) nano clay			Nur Humairah and Zuraida (2012)
Cassava/agar	Zinc Oxide		Film casting	Mahuwala <i>et al.</i> (2020)
Tapioca	Minor gums	-		Kim <i>et al.</i> (2015)
	CMC	Turmeric oil		Mustapha <i>et al.</i> (2019)
Cassava	Woven-bamboo fiber	Epoxidized waste cooking oil Lime juice		Silviana and Dzulkarom (2019)
	Carrageenan and nano clay	Glycerol	Film extrusion	Suryanto <i>et al.</i> (2020)
	Linear low-density polyethylene (LLDPE)		Hydraulic pressing	Irwanto <i>et al.</i> (2020)
Potato	Polypropylene (PP)		Milling	Roy <i>et al.</i> (2015)
Arrowroot	Polycaprolactone (PCL)	Polyethylene glycol	Nonsolvent-induced separation (NIPS)	Damian <i>et al.</i> (2020)
Sugar palm	Sugar-palm nanocrystalline cellulose (SPNCC)	Sorbitol and glycerol Cinnamon oil as additives		Syafiq <i>et al.</i> (2021)
Rice	PVA	Glycerol		Parvin <i>et al.</i> (2010)
Corn	PVA	Glycerol		Pinar Terzioglu and Par (2020)
	Lemon peel	Citric acid		Romero-Bastida <i>et al.</i> (2015)
Cassava	MMT		Solution casting	Kargarzadeh <i>et al.</i> (2017)
Potato	Rusk husk cellulose	Glycerol		Cyras <i>et al.</i> (2008)
PLA	Palm kernel shell	Butyl methyl acrylate (BMA)		Hasnan <i>et al.</i> (2016)
Tapioca	Carrageenan, nano chitosan suspension	Glycerol		Rochima <i>et al.</i> (2018)
Corn	Rice straw	Lemon juice		Noor Zulaika <i>et al.</i> (2020)
	Soy protein isolate	Glycerol		Mansoori <i>et al.</i> (2019)
Tapioca	Graphene oxide			Maran <i>et al.</i> (2014)
Sago	Agar			Fath <i>et al.</i> (2019)
Tapioca	Pectin from orange peel		Solution casting technique with ultrasonic wave treatment	Mochamad <i>et al.</i> (2020)
Cassava	Sugarcane bagasse	Glycerol		Suryanto <i>et al.</i> (2019)
PLA	MMT nano clay		Solvent casting	Anuar <i>et al.</i> (2017)
	Durian skin fiber (DSF)	Chloroform		

chains in the liquid medium as it emits ultrasonic waves and produces cavitation that will bump with each other to interrupt the bond therefore can be dispersed well in the tapioca matrix. The same treatment is also done by Suryanto *et al.* (2019) in which nano clays are uniformly dispersed in the cassava matrix as the agglomeration of nano clays is prevented when ultrasonic waves are emitted. Meanwhile, some researchers used citric acids, yerba mate, epoxidized waste cooking oil, and lemon juices as crosslinkers as they can combine the

amylopectin and amylose in the starch thru hydrogen bonding as they possess a high number of hydroxyl groups (Yu *et al.*, 2013; Kargarzadeh *et al.*, 2017; Mustapha *et al.*, 2019; Beigmohammadi *et al.*, 2020; Noor Zulaika *et al.*, 2020). As bonding between the molecules occurs, tensile strength can be improved.

3. Film characterization

The characterization analyses are classified based on properties that are evaluated to be used in packaging.

The properties of the films include morphological, optical, thermal, barrier, mechanical, chemical composition, biodegradability, physicochemical properties, and antimicrobial properties.

3.1 Morphological and chemical properties

For the characterisation analyses, morphological properties of the films are vital in determining the type of biocomposite that can be utilized for packaging depending on its surface characteristics. The surface characteristics will define the packaging quality. Reinforced starch biocomposites have better morphological properties as the fillers help in the plasticization of the starch thus producing a good surface property (Reinaldo *et al.*, 2021). Morphological properties can be characterized by the dispersion and homogeneity of the reinforcement within the starch matrix with the use of scanning electron microscopy (SEM) analysis, in which the particles of the filler can be magnified to a visible scale (Silviana and Dzulkarom, 2019; Reinaldo *et al.*, 2021). In the morphological analysis, focused electrons are beamed over the sample surface, and the electrons will interact with the elements present in the sample, thus producing various signals. The signals are then scanned by the SEM, which will produce an image based on the signals that represent the data on the surface topography and composition. The magnification of the image can reach up to 1,000,000× in the voltage of 10.00 kV (Suryanto *et al.*, 2020).

Based on Table 2, for SEM analysis, good adhesion and dispersion of the fillers in the starch matrix will produce a homogenous and uniform film that can improve the mechanical properties of the film. Good dispersion is achieved when the starch matrix can be seen to be completely covering the filler in SEM (Johar and Ahmad, 2012). Instead, the existence of pores means improved water absorption due to the hydrogen bonding of the matrix due to the nature of hydrophilic starch (Damian *et al.*, 2020). Thus, as starch content increases, the hydrophilicity will increase too, producing a more porous surface of the film. On the other hand, the rough surface of the starch and CNC biocomposite is similar to the surface of the native starch matrix this is because CNC as reinforcement will not alter the physical appearance of the film making it similar to the native starch films therefore, CNC has no contributing effect on the morphological properties of the biocomposite film (Johar and Ahmad, 2012). Meanwhile, the rough surface of the cassava and rice husks bran biocomposite film is due to the weak interfacial adhesion between filler and matrix this is due to the big size of the filler particles that can cause roughness of the dispersion (Kargarzadeh *et al.*, 2017). Homogeneity between the starch particles and filler is wanted as it will produce a smooth film which

will be desired for good packaging.

On the other hand, the crystallinity structure of the starch granules as morphological properties can be observed with x-ray diffractometry (X-RD) analysis in which the crystallinity index can be obtained (McGlashan and Halley, 2003; Anuar *et al.*, 2017). As starch can be crystallized, the crystallinity will determine the surface properties of the film. Thus, X-RD provides information of the crystalline structure by scattering the X-rays beams on the surface sample, the rays are then diffracted producing a diffraction angle (Kumar *et al.*, 2013). The diffraction angles will describe the certain spacing between atoms or crystals in the sample, hence, the greater the angle, the higher the crystallinity of the sample. The reinforcement of MMT in the potato-starch matrix is found to increase the crystallinity content due to the extremely constructed MMT that could hold its structure despite being immersed in gelatinized starch dispersions (Romero-Bastida *et al.*, 2015). A similar finding is found by (Mukurubira *et al.*, 2017) who also reported in which the reinforcement of amadumbe starch into the potato-starch biocomposite has increased the crystallinity as starch is naturally rich in the crystalline structure, therefore, increasing the crystallinity of the film thus providing better tensile strength and stiffness.

Conversely, in terms of the chemical composition of the film, the use of Fourier Transform Infrared Spectrometry (FTIR) analysis can help to detect the material composition and structure by characterizing the functional groups and chemical bonding present. FTIR utilizes infrared radiation, in which the rays will be radiated on the sample, and the interaction between the matter and radiation will produce spectrum data of absorption that will be analyzed to differentiate and identify each compound that is present in the sample. FTIR is considered a fingerprint technique this is because each compound will have specific spectrum data, therefore the spectrum data from the sample can be compared with the data from previous experiments for identification purposes (Holler and Crouch, 2013). FTIR is commonly employed to show the interaction of chemical bonding between the starch and fillers upon fabrication process. The band wavenumbers above 1500 cm^{-1} represent the functional groups that are present in the matrix of the biocomposites films as wavenumber 1609 cm^{-1} represents the presence of yerba mate phenolic compound in potato starch, PVA and yerba mate biocomposite film (López-córdoba *et al.*, 2019). Additionally, wavenumber 1653 cm^{-1} also represents the keratin compound in cornstarch, chemically modified starch, and keratin biocomposite film (Bodirlau *et al.*, 2013). Meanwhile, wavenumbers above 3000 cm^{-1} represent the hydroxyl group interaction in which the

Table 2. Morphological properties and chemical composition.

Type of starch	Reinforcement/filler	Morphological properties		Chemical compositions		References
		Surface texture	Wavelength (cm ⁻¹)	Description of FTIR/Assignment		
Arrowroot	Polycaprolactone (PCL)	Homogenous and porous surface.	3438-3439.5	The appearance of OH group peak.		Damian <i>et al.</i> (2020)
Cassava	Nano clay	Nano clay uniformly dispersed.	1400-1600	Formation of C=C bond.		Suryanto <i>et al.</i> (2019)
	MMT + cinnamaldehyde	Smooth with no bubbles.	1600-1700	The vibration of the aromatic rings.		Cristina <i>et al.</i> (2013)
Corn	Soy and Graphene oxide	The film is smooth and homogenously dispersed.	3282	New intramolecular hydrogen bonds between functional group of the starch matrix and oxygenic groups of nanoplatelets.		Mansoori <i>et al.</i> (2019)
	Organoclay + lactic acid	Presence of micropores and nano cracks.	3354	O-H stretching vibrations.		Salimi <i>et al.</i> (2017)
	Chemically modified starch + keratin	CMS was well-dispersed between the matrix without obvious aggregation.	1653	Absorption of hydroxyl groups.		Bodirlau <i>et al.</i> (2013)
Potato	Chestnut husks	Uniform dispersion of filler.	3200-3600	O-H stretching bands.		Torres <i>et al.</i> (2019)
	PVA and Yerba mate	Rough with visible small cracks.	1609	Stretching of aromatic rings due to the presence of yerba mate phenolic compound.		López-córdoba <i>et al.</i> (2019)
	MMT	MMT is poorly dispersed and form aggregates of MMT on the film.	3530	Hydroxyl groups that participated in hydrogen bonding formation.		Cyras <i>et al.</i> (2008)
Rye flour	Microcrystalline cellulose	Uniform distribution of the filler.	3289	NA		Beigmohammadi <i>et al.</i> (2020)
Sago	Rattan NCC	The film looks quite well and evenly mixed.	3287	Presence of O-H group.		Fath <i>et al.</i> (2019)
	Nano clay	Less clay content in the film produces a homogenous phase and well-dispersed clay platelet in the starch matrix.	3273.31-1642.66	Bonding of O-H groups between starch and nano clay (due to the bound water present in the starch).		Nur Humairah and Zuraida (2012)
Sugar palm starch	SPNCC + cinnamon essential oil	Smooth and uniform.	3266	Presence of EO that contains hydrocarbons in the matrix.		Syafiq <i>et al.</i> (2021)
	Sugar palm cellulose	Rough and non-noticeable clusters of SPC.	3267	Correspond to O-H group.		Sanyang <i>et al.</i> (2016)
	Rice straw cellulose	Smooth surface.	3287.45 -3289.06	Stretching of O-H in cellulose.		Noor Zulaika <i>et al.</i> (2020)
Thermoplastic corn (TPC)	Bacterial cellulose	Homogenous with the presence of flakes.	3300-3600	To show that there is an interfacial adhesion between starch matrix and filler.		Santos and Spinacé (2021)

starches and the fillers are bonded through the hydroxyl group therefore is described by the stretching vibrations of the O-H (Nur Humairah and Zuraida, 2012). The wavenumbers act as fingerprints in which the presence of compounds can be proven as stated in Table 2. The analysis is simple, fast, and efficient which can be used to observe the morphological properties of the starch biocomposite films.

3.2 Optical properties

3.2.1 Colour and transparency

Films made from pure starch are usually colourless and therefore transparent. However, upon reinforcement, the colour and transparency might change depending on the type of fillers (Çokaygil *et al.*, 2014; Suryanto *et al.*, 2020). In native tapioca starch film, the transparency of the film is found to be 18.80 which is the most

transparent film (Othman *et al.*, 2020). The colour of the films is usually measured with a chromameter and is compared with the Hunter lab colour scale to validate the colour (Rochima *et al.*, 2018). Meanwhile, transparency is measured with the UV-Visible spectrometer in which the transparency will be depending on the thickness of the packaging (Podshivalov *et al.*, 2017). In packaging, partial transparency is an advantageous characteristic that enables consumers to verify the quality and condition of the product, however high opacity is also desirable as it can protect the product from UV rays (Santos and Spinacé, 2021). When the tapioca-starch film is reinforced with Gellan gum, the transparency declined to 1.67 which still can be considered transparent (Kim *et al.*, 2015). High transparency can be achieved when there's no aggregation of fillers in the starch matrix (Podshivalov *et al.*, 2017).

3.2.2 Thickness

Other than the thickness that has been standardized for certain experiments, the thickness of the biocomposite films is varied as it depends on the intrinsic association between the starch matrix and the fillers (Kim *et al.*, 2015). The clusters of fillers that can form in the starch matrix can affect the thickness variations (Syafiq *et al.*, 2021). In starch and MMT biocomposite, the thickness increases upon reinforcement due to the large size of the nano clay particles (Nur Humairah and Zuraida, 2012). With reinforcement, the thickness of the films will increase. Moreover, the fabrication method can also influence the thickness of the films in which the film blowing method can produce the thinnest film ranging from 0.007 mm to 0.125 mm (Mangaraj *et al.*, 2019). The thickness range for the film casting method is usually between 0.02 – 0.10 mm (Yu *et al.*, 2013). Moreover, the corn starch with organoclay and lactic acid biocomposite film has the highest tensile strength of 82.4 MPa with a thickness of 0.152 mm (Salimi *et al.*, 2017). Thus, thickness also influences tensile strength.

3.2.3 Density

Density is an important property since a high density contributes to high transport costs and affects mechanical characteristics (Syafiq *et al.*, 2021). The film that possesses the lowest density is sago-starch, pectin, and nanocrystalline cellulose biocomposite at 0.068 g/cm³ while the highest density is the sago-starch and nano clay biocomposite at 1.49 g/cm³. The high density is due to the high percentage of nano clay at 15% for weight fraction as clay is relatively denser than cellulose meanwhile the nanocrystalline cellulose is effectively dispersed without any agglomeration that makes it less dense (Agustin *et al.*, 2013; Irwanto *et al.*, 2020). In

starch and MMT biocomposite film, as the filler increases, the spacing within the matrix becomes nearer therefore the empty spaces are lessened, making the film denser (Nur Humairah and Zuraida, 2012).

3.3 Mechanical properties

3.3.1 Tensile strength

Tensile strength describes the maximum forces that a material can bear upon breaking. The tensile strength can be measured with a texture analyzer where the film will be stretched (Silviana and Dzulkarom, 2019). Tensile strength reflects the ability of composite material to bear stress or load without damage or injury expressed with maximal tension before breakage and is the absolute tensile strength (Suryanto *et al.*, 2019). Factors that can affect tensile strength also include the matrix material compatibility with the fillers, the homogeneity dispersion of the fillers in the matrix, the relative ratio of matrix and filler, and the materials that made up the matrix in the biocomposite (Suryanto *et al.*, 2020). The fabrication environment, the type of fillers, and their orientation also ruled the tensile strength (Vinod *et al.*, 2020). Tensile strength analysis aims to determine whether the film is strong enough to be utilized for packaging in terms of its material strength to bear weight and force. Based on Table 3, the biocomposite of corn starch and organoclay with lactic acid possessed the highest tensile strength, this is because there is a compatibility between the starch matrix and the filler as their hydrophilic groups interacted in a high value of interfacial adhesion, therefore, improving the mechanical properties (Salimi *et al.*, 2017). The strong interfacial adhesion between starch and cellulose matrix in the tapioca and rice straw cellulose biocomposite also enables an efficient stress transfer of the film thus obtaining high tensile strength (Noor Zulaika *et al.*, 2020). On the contrary, the cassava and acerola residues biocomposite possesses the lowest tensile strength which can be due to the high content of sugar of the acerola residues which gives plasticizing effect, therefore, reducing its glass transition temperature which decreases the modulus of elasticity and tensile strength (Reinaldo *et al.*, 2021).

3.3.2 Young's modulus

In addition to tensile strength for mechanical properties, Young's modulus is a reliable indicator of the film's stiffness and durability (Othman *et al.*, 2020). Young's modulus defines the ability of the film to return to its original form after the load is removed, in which low modulus indicates high elasticity and vice versa (Helmenstine, 2019). Based on Table 3, thermoplastic corn starch with organoclay biocomposite possesses the highest young's modulus value as well as its tensile strength as films that possess high modulus signify that

Table 3. Optical, barrier and mechanical properties

Type of starch	Reinforcement/ filler	Optical Property	Barrier property	Mechanical properties			References
		Thickness	WVP ($\times 10^{-10}$ g/ms Pa)	Tensile strength (MPa)	Young's modulus (MPa)	Elongation at break (%)	
Amadumbe	SNC	0.204	NA	8.110	NA	NA	Mukurubira <i>et al.</i> (2017)
Arrowroot	PCL	0.030	NA	3.407	1.894	180.929	Damian <i>et al.</i> (2020)
	Acerola residue	NA	NA	0.500	0.800	458.000	Reinaldo <i>et al.</i> (2021)
	Carrageenan and nano clay	NA	NA	2.480	21.220	17.910	Suryanto <i>et al.</i> (2020)
	CMC	NA	NA	15.000	NA	NA	Mustapha <i>et al.</i> (2019)
Cassava	Bamboo fiber	NA	NA	38.490	NA	NA	Silviana and Dzulkarom (2019)
	Nano clay	0.153	NA	28.010	100.400	27.900	Suryanto <i>et al.</i> (2019)
	CMC	0.017	NA	15.340	NA	NA	Fatin <i>et al.</i> (2018)
	Rice husks CNC	0.300	NA	3.800	45.000	85.000	Kargarzadeh <i>et al.</i> (2017)
	Rice husks	0.300	NA	6.300	261.700	NA	Johar and Ahmad (2012)
	PVC	0.290	NA	25.110	52.700	275.190	Pinar Terzioglu and Par (2020)
	Soy and Graphene oxide	0.170	12.380	5.900	449.300	11.830	Mansoori <i>et al.</i> (2019)
Corn	MMT	NA	0.900	18.000	1650.000	2.000	Romero-Bastida <i>et al.</i> (2015)
	Organoclay + lactic acid	0.152	3.200	82.400	531.000	5.200	Salimi <i>et al.</i> (2017)
	Garlic stalk cellulose	0.200	NA	15.600	439.600	NA	Agustin <i>et al.</i> (2013)
	Chemically modified starch + keratin	0.200	NA	25.300	1645.000	4.500	Bodirlau <i>et al.</i> (2013)
	Rice bran	0.079	6.350	16.000	663.000	3.100	Cano <i>et al.</i> (2014)
Potato	Chestnut husks	NA	NA	4.850	0.250	14.430	Torres <i>et al.</i> (2019)
	PVA and Yerba mate	0.100	4.040	2.400	37.600	38.000	López-córdoba <i>et al.</i> (2019)
	Jute fiber	2.500	NA	10.46	NA	NA	Verma <i>et al.</i> (2018)
	MMT	0.250	NA	5.200	195.6	46.80	Cyras <i>et al.</i> (2008)
Rye flour	Microcrystalline cellulose	1.000	8.700	1.340	64.02	6.610	Beigmohammadi <i>et al.</i> (2020)
Sago	Nano clay	0.458	NA	NA	NA	NA	Nur Humairah and Zuraida (2012)
Sugar palm starch	SPNCC	0.253	NA	5.300	130.520	13.990	Syafiq <i>et al.</i> (2021)
	Sugar palm cellulose	NA	1.854	7.790	20.110	32.800	Sanyang <i>et al.</i> (2016)
Tapioca	Banana pseudo stem	0.100	1.060	24.200	659.690	111.710	Othman <i>et al.</i> (2020)
Thermoplastic corn (TPC)	Bacterial cellulose	0.270	NA	11.230	0.430E+11	7.380	Santos and Spinacé (2021)

they have high tensile strength or a low elasticity characteristic (Suryanto *et al.*, 2020). For corn-starch and MMT film, the high modulus can be explained by the fair dispersion of the filler in the matrix, therefore, achieving an effective interfacial adhesion (Saber *et al.*, 2017; Santos and Spinacé, 2021). Furthermore, if the fillers develop big clusters in the starch matrix and interfere with the starch crystallization, the modulus will decrease (Wypych, 2016). Thus, the compatibility of the starch and fillers as biocomposites can also be determined with young's modulus. Additionally, the film crystallinity is also represented by the modulus, as high crystalline structures also have high modulus values (Kargarzadeh *et al.*, 2017). Hence, the elasticity of the film depends on the type of fillers, sizes, and dispersion in the starch matrix.

3.3.3 Elongation at break

The elongation at break is the full change in film length while the tensile strength is achieved before the film breaks relative to the original length (Suryanto *et al.*, 2020). The elongation at break is measured by rationing the final length and initial length of the film after fracturing in percentage. The elongation at break is mostly inversely proportional to the tensile strength which might be due to the decline in the ability to adjust the structure of the bonding between the matrix and the filler. The tension is usually a measure of the inner bond between the filler and the matrix. If the relationship between the two is weak, the interaction would be weak (Kargarzadeh *et al.*, 2017). As the elongation of break decreases, the biocomposite becomes stiffer and harder to break this is due to the decline of molecular mobility of the starch due to the presence of fillers (Sanyang *et al.*, 2016). Moreover, the decrement in elongation at break upon reinforcement is common in the majority of the biocomposites as the tensile strength increases as the ability of the film to withstand pressure without any formation of ruptures (Wypych, 2016).

3.4 Thermal barrier properties

3.4.1 Thermogravimetric analysis

The thermogravimetric analysis (TGA) is the analysis that measures the mass sample changes when the temperature is shifted. The effect of the reinforcement of the starch matrix on the thermal barrier stability can be determined by TGA as the heat resistance can be analyzed by determining the degradation temperature of the matter (Cyras *et al.*, 2008; Scheibe *et al.*, 2014). The corn starch and chemically modified starch with keratin biocomposite are 85% degraded at 600°C (Bodirlau *et al.*, 2013). However, for cassava/agar and Ag Oxide biocomposite, it is 74.74% degraded at 400°C (Mahuwala *et al.*, 2020).

This can be due to the fair dispersion of the filler in the starch matrix thus increasing thermal stability (Johar and Ahmad, 2012). Cyras *et al.* (2008) also concluded that the reinforcement of MMT improves the overall thermal stability of the biocomposite of potato starch as MMT functions as a heat barrier. The degradation temperature represents the thermal stability of the chemical and physical bonding between the fillers and starch matrix (Mahuwala *et al.*, 2020). Therefore, upon reinforcement, the thermal stability of starch biocomposite increased. It is advantageous for biocomposite films to possess high thermal stability to protect the product inside (Marie Arockianathan *et al.*, 2012).

3.4.2 Differential scanning calorimetry analysis

On the other hand, differential scanning calorimetry analysis can be used to determine the glass transition and melting temperature of the film. DSC is a thermoanalytical analysis in which the thermal changes are measured producing crystallization and melting curves. The temperature at which the amorphous starch structure turns to a thermoplastic is called glass transition temperature in which the carbon chains begin to shift. Each compound when heated will take different times for the shifting of the carbon chain to occur depending on the stability of the chemical bonds, therefore the lower the glass transition temperature, the more it resembles conventional plastics (Gopal, 2014). The Tg for cassava-starch biocomposite is found to be shifted to a low temperature at 57°C as the chestnut husks are reinforced into the matrix (Kargarzadeh *et al.*, 2017). Similarly, when potato starch is reinforced with chestnut husk, the Tg decreased as the filler content increases demonstrating that the segmental mobility of starch molecules has improved (Torres *et al.*, 2019). Hence, the low Tg is caused by the increase in mobility of the starch polymers, therefore, reducing the intermolecular forces thus permeability and processability of the films are improved (Mochamad *et al.*, 2020). Tg is influenced by the individual molecular weight of the elements, the intermolecular forces between the starch and fillers, and the plasticizers used in the fabrication (Mangaraj *et al.*, 2019).

Conversely, melting temperature (Tm) is the temperature where the crystalline structure of the starch becomes a disordered liquid structure in which thermal energy is needed to free the polymer molecules of starch from the crystalline structure. The Tm is also analyzed with DSC in which the melting curve is produced upon melting of the samples (Das *et al.*, 2011). If the Tm increased, more thermal energy is needed wherein the biocomposite of corn-starch and MMT, upon reinforcement, the melting temperature increased to

180.59°C from 164.39°C as MMT facilitates the structure of a bigger crystal domain and reduces the movement of the polymer chains (Romero-Bastida *et al.*, 2015). Similarly, the T_m in amadumbe starch reinforced with starch nanocrystals films also increases due to the higher crystallinity of the biocomposite (Mukurubira *et al.*, 2017). A high melting point indicates that the film can withstand high temperatures.

3.5 Barrier properties

3.5.1 Wettability

Wettability determines the hydrophobicity of the film in which the contact angle formed when water is dropped on the surface is calculated (Scheibe *et al.*, 2014). Cassava starch film without reinforcement had a lower than 90° contact angle in 120 s as starch in nature is hydrophilic (Mustapha *et al.*, 2019). However, the wettability of cassava and CMC biocomposite is found to be improved with a contact angle higher than 90° in 120 s (Mustapha *et al.*, 2019). Similarly, PLA and palm kernel shell biocomposite also possess high contact angles as the interfacial interaction of the matrix is improved therefore increasing the hydrophobicity of the film (Hasnan *et al.*, 2016). A contact angle higher than 90° describes that the surface is not easily wettable, thus having a good water-resistant property (Narkchamnan and Sakdaronnarong, 2013). Furthermore, the wettability of the film can be improved when the starch matrix forms a good interfacial adhesion with the filler thus increasing the hydrophobicity.

3.5.2 Moisture content

Moisture content indicates the percentage of water content in the films based on their weight. Thus, low MC is important for the barrier properties so that there will be no moisture exchange between the packaging and the product (Mukurubira *et al.*, 2017). This is because the presence of water can interrupt the mechanical properties of the film as it is closely related to the relative humidity in which the exchange of moisture between the film and the product can occur (Xie *et al.*, 2015). Mahuwala *et al.* (2020) found that when cassava is reinforced with agar and Ag Oxide, the MC of the film was found to be lower at 11.64% from 34.44% due to the metal nanoparticles that are hydrophobic as fillers. Similarly, López-córdoba *et al.* (2019) also found that PVA and yerba mate as fillers in the potato-starch matrix lowering the MC by 12% from 15.2%. As starch in nature is sensitive to water, it possesses high moisture content if solely used as films, therefore, with reinforcement, the moisture content can be decreased as the ability of the films to absorb water also decreases (Mangaraj *et al.*, 2019).

3.5.3 Water vapour transmission rate

Water vapour transmission rate (WVTR) is analyzed so that the in and out water vapour transfer of the product can be delayed as it represents the amount of water that infuses within materials of specific thickness for each unit duration and region at a given permeant gradient level (Çokaygil *et al.*, 2014). Two factors need to be considered in determining WVTR which are relative humidity and temperature in which when these two factors are high, the WVTR will also increase (Basha *et al.*, 2011; Othman *et al.*, 2020). The WVTR for corn-starch, soy, and graphene oxide biocomposite is recorded at 8.84 g/m² h (Mansoori *et al.*, 2019). The result is almost similar to sago-starch and rattan NCC biocomposite at 9.0 g/m² h in which the fillers act as barriers that slow down the vapour transfusion (Al Fath *et al.*, 2019). WVTR must be low that there is no vapour transmission between the packaging and the product therefore, the quality of the product is protected. Low WVTR indicates that the film has good humidity protection in which it can resist moisture input or output (Galić *et al.*, 2017).

3.5.4 Water vapour permeability

Water vapour permeability (WVP) is derived from WVTR, in which WVP is calculated by standardizing WVTR with the thickness of the films and the difference in water vapour partial pressure (Basha *et al.*, 2011). WVP must always be low to avoid moisture from surrounding to be transferred into the product (Mukurubira *et al.*, 2017). Romero-Bastida *et al.*, (2015) found that MMT as fillers lessens the permeability of the corn-starch matrix thus low WVP is achieved. Similarly, Othman *et al.*, (2020) found that as tapioca is reinforced with banana pseudostem cellulose, the WVP is lowered as a rigid hydrogen bond network of cellulose is formed in the biocomposite.

3.6 Biodegradability

Biodegradation is when the structure of the film is degraded along with organic wastes in the presence of microorganisms and released back to enrich the soil (Faris *et al.*, 2014). It can be expressed in terms of the biodegradation rate at which soil burial test analysis can be performed. In soil burial tests, sugar from starch is utilized by the microorganisms as the source of energy thus degradation of the starch polymers occurs. The biodegradation rate is found to be increasing along with the starch content in the biocomposite films of arrowroot starch and PCL at 2.29% after 15 days, and the biocomposite film of corn-starch, PLA, and lactic acid also increased with the starch content (Mansoori *et al.*, 2019; Silviana and Dzulkarom, 2019). Therefore, the

higher the starch content in the biocomposite film, the higher the biodegradation rate. Furthermore, Roy *et al.* (2015) also reported that the biodegradation rate of potato-starch and polypropylene film increased to 10.6% after 120 days. The authors also describe the stages involved in the degradation process of the film which are composting and curing phases. In the composting phase, the degradation process caused an increment in temperature which caused a strong microbial activity due to the presence of oxygen. However, in the curing phase, as no oxygen is longer available, the film is still degraded at a lower rate. Therefore, the biocomposite will still be degraded over time.

3.7 Antimicrobial activity

Additives are added to enhance the antimicrobial activity of the packaging that can help to preserve the quality of the product, especially food. Additives as antimicrobial agents can defer and hinder microbial growth that can cause spoilage (Majhi *et al.*, 2020). Additives can be incorporated into the packaging as bioactive compounds in which cinnamon EO when used as an additive is found to be inhibiting the growth of *Bacillus subtilis* and *Escherichia coli* effectively (Anuar *et al.*, 2017). Similarly, in the biocomposite of sugar palm starch and sugar palm nanocellulose, the incorporation of cinnamon EO has an inhibitory effect against *Bacillus subtilis* and *Escherichia coli*, and *Staphylococcus aureus* where the antimicrobial element in cinnamon EO is capable of disrupting the cell membrane of the bacteria (Syafiq *et al.*, 2021). Furthermore, nano chitosan suspension when used as filler in tapioca and carrageenan biocomposite film also exhibited antimicrobial activity against *Escherichia coli* and *Staphylococcus aureus* (Rochima *et al.*, 2018). Additionally, AgNO₃ is also utilized as an antimicrobial element in the starch /agar biocomposite which possesses the highest antimicrobial activity toward *Pseudomonas aeruginosa* which is a Gram-negative bacteria (Mahuwala *et al.*, 2020). Furthermore, benzoate as an antimicrobial agent in arrowroot starch film exhibited the biggest inhibition zone against *Staphylococcus aureus* and *Salmonella* (Ratnawati and Afifah, 2019).

4. Application in packaging

For the analyses outcomes to be applied in packaging, the characteristics of the films depend on the purpose of packaging which is the product. In general, the fillers must be homogeneously dispersed in the starch matrix for better mechanical properties which is also supported by FTIR analysis that shows hydroxyl bonding to prove the chemical bonding between the starch and fillers. A chemically bonded biocomposite will possess a high interfacial adhesion, therefore, will have improved

mechanical and morphological properties compared to native starch films. Films that possess high crystallinity also acquire better mechanical properties (Li *et al.*, 2015). The colour and transparency of the films will be depending on the purpose of the product. If transparent, the product is visible. If opaque, the product is protected from UV-rays. The same goes for thickness in which thick film can protect the product while thin film makes the product visible. Most importantly, tensile strength, young's modulus, Eb, thermal degradation temperature, and melting temperature must be high to produce the best biocomposite film in terms of mechanical, and thermal properties (Mangaraj *et al.*, 2019). However, moisture content, density, WVP, WVTR, and glass transition temperature must be low so that the film is light, dry, and easy to process for packaging (Rastogi and Samyn, 2015; Mustapha *et al.*, 2019). These barrier properties are important to be considered, as they can affect the quality of the packaging. The biodegradability rate of the film must also be high so that when disposed of on landfills, it can be absorbed back into the soil (Li *et al.*, 2015). Biodegradability is the main factor for the packaging to be considered as green packaging. Lastly, additives used as antimicrobial agents can fully function as active packaging (Balan *et al.*, 2015). Active packaging can protect the product against bacteria that can cause spoilage which can jeopardize the quality of the products, especially food.

Starch-based biocomposite films have been in several types of packaging including food paper coating packaging (Fatin *et al.*, 2018), antibacterial food packaging (Anuar *et al.*, 2017), antioxidant packaging (Reinaldo *et al.*, 2021), fruit coating (Saber *et al.*, 2017), and shopping and garbage bags (Parvin *et al.*, 2010). The reinforced films have exhibited better morphological properties (Nur Humairah and Zuraida, 2012; Fatin *et al.*, 2018), improved mechanical properties (Romero-Bastida *et al.*, 2015; Salimi *et al.*, 2017; Pinar Terzioglu and Par, 2020; Santos and Spinacé, 2021; Syafiq *et al.*, 2021), enhanced barrier properties (Mukurubira *et al.*, 2017; Salimi *et al.*, 2017) and upgraded biodegradability (Roy *et al.*, 2015).

5. Conclusion

In conclusion, the abundance of starch in nature has enabled them to be utilized in packaging as films. The weakness and brittleness of starch-based films can be overcome with reinforcement. When starch is reinforced with fillers, they become biocomposite films. There are several fabrication processes such as moulding, extrusion, and casting in which glycerol is often employed as the plasticizer. Characterization analyses based on the films' properties such as morphological,

optical, mechanical, barrier, thermal barrier, and biodegradability have been reviewed in which reinforcements are found to be improving these properties. For morphological properties, they can be characterized by FTIR, SEM, and X-RD analyses to show the functional groups that are present in films, the image of the surface film at high magnification, and the crystallinity of the starch in the film.

Meanwhile, for mechanical properties, they can be characterized by tensile strength, young's modulus, and elongation at break, in which most, upon reinforcement, tensile strength, and young's modulus will increase as they represent the film durability while elongation at break will decrease as the films will become inelastic. Moreover, thermogravimetric and DSC analyses are employed to analyze the thermal stability, crystallization and melting point of the films. Similarly, the barrier properties of films are described by moisture content, WVTR, and WVP which must be always low so that the films for packaging are not easily wettable.

Most importantly, biocomposite films are biodegradable in which they can be decomposed into the environment upon reaching the landfill and thus will not contribute to pollution. Therefore, with different reinforcements, various outcomes of those properties can be used for distinctive purposes of packaging which include paper coating, active packaging, and edible food coating. Reinforcement of starch with various fillers as biocomposite films for packaging can be a stepping stone in reducing the number of plastics and therefore can contribute to a greener environment.

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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