

Fruit Waste Valorization for Biodegradable Biocomposite Applications: A Review

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Currently, food waste is a major concern for companies, governments, and consumers. One of the largest sources of food waste occurs during industrial processing, where substantial by-products are generated. Fruit processing creates a lot of these by-products, from undesirable or “ugly fruit,” to the skins, seeds, and fleshy parts of the fruits. These by-products compose up to 30% of the initial mass of fruit processed. Millions of tons of fruit wastes are generated globally from spoilage and industrial by-products, so it is essential to find alternative uses for fruit wastes to increase their value. This goal can be accomplished by processing fruit waste into fillers and incorporating them into polymeric materials. This review summarizes recent developments in technologies to incorporate fruit wastes from sources such as grape, apple, olive, banana, coconut, pineapple, and others into polymer matrices to create green composites or films. Various surface treatments of biofillers/fibers are also discussed; these treatments increase the adhesion and applicability of the fillers with various bioplastics. Lastly, a comprehensive review of sustainable and biodegradable biocomposites is presented.

Keywords: Fruit waste; Biodegradable; Biobased plastics; Biocomposites

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INTRODUCTION

One of the biggest challenges facing the world today is to produce enough food to feed the rapidly growing population. Despite efforts to produce enough food, much of it is wasted before it even reaches the consumers. According to the Food and Agriculture Organization (FAO) of the United Nations, approximately 1.3 billion tonnes of food is lost every year (Gustavsson *et al.* 2011). Fruit and vegetable losses in industrialized nations can exceed 32% before reaching the point of sale (Gustavsson *et al.* 2011). A prominent solution to reduce food loss is to find alternative uses for products that are rejected by consumers or leftovers from food processing (ASTM D6400-04 2004). Research relating to food waste has been ongoing since the 1990s (Kroyer 1995). To assess the feasibility and prepare to utilize these by-products in industry, it is essential to identify, quantify, characterize, and analyze the by-products of choice (Rosentrater 2004). Full utilization of the by-products can only be realized when the materials' information is available.

One way to increase the use of food waste is to process it into filler and then

incorporate it into a polymer matrix to provide strength and decrease the amount of polymer required in the composite. A further benefit to this approach is the reduction in material cost, as the more expensive polymer is replaced with low-cost filler. By combining different fruit waste fillers with a polymer, composite systems of unique properties can be tailored for specific applications. Common properties of interest include strength, toughness, and thermal stability, among others. These composites can be classified as biocomposites because they contain a biologically derived resource, *i.e.*, the natural filler. They can be considered biodegradable if a biodegradable polymer matrix is used. In this article the term “biocomposite” will be used when both the polymer matrix and the filler particles both come from plant sources. Materials with a petrochemical-based matrix and any kind of reinforcement, including plant-based, will just be called “composites”.

“Biodegradability” is an all-encompassing term used to describe a material’s ability to break down. However, a subsection within biodegradability is compostability. Compostable materials have the ability to be degraded into benign substances under certain conditions, as defined within internationally recognized standards. Factors that affect the degradability of a sample include its size, composition, and thickness, among other characteristics (ASTM D6400-04 2004). Biodegradable or compostable plastics have less environmental impact compared to non-biodegradable or non-compostable plastics because they do not place such a heavy burden on the environment. The biodegradable, compostable plastics contribute to a circular economy in which the products at the end of their life return to the soil and new starting material begins again. There is an increasing demand for these types of products, as consumers are becoming more aware of the impact of non-biodegradable plastics on the environment. Compostable alternatives can potentially alleviate disposal concerns and help create sustainable, environmentally friendly products.

This work is instrumental in providing information to both consumers and industry on the availability of sustainable polymers and composites. The cost benefits associated with the use of natural fillers and sustainability are among the many benefits to using these materials. This work discusses the availability of fruit waste materials around the world. Often, these materials can be obtained at little to no cost, as the natural fillers generated from fruit waste have limited purpose to date. The circular economy, as discussed in this work, focuses on the regeneration of value-added products from previously used materials (Ellen MacArthur Foundation 2017). A circular approach is far more sustainable in the long run, as compared to the linear practices many companies follow today, which require resources to make products and dispose of products at the end of life. Thus, the use of biocomposites is extremely important going forward, especially in single-use plastic applications (Ellen MacArthur Foundation 2017).

Biopolymers can be classified into different categories, depending on how they are produced, whether or not they are biodegradable, and their structures. There are currently many different types of biopolymers available on the market. The major considerations for classification of biopolymers are the source of the starting material and its biodegradability. Recently there has been research on the combination of petroleum-based or biobased polymers with natural fillers or fibers, which may or may not be biodegradable. It is important to clarify that although a material may be biologically based, it might not be biodegradable. This paper intends to focus on biodegradable polymers, regardless of their starting material.

There have been some studies examining composites derived from natural fillers

or fibers and non-biodegradable polymers, which are more sustainable than entirely petro-based materials. These studies feature the combination of fruit waste with petroleum-based polymers such as high density polyethylene (HDPE) (Banat and Fares 2015; Satapathy and Kothapalli 2018), polypropylene (PP) (Naghmouchi *et al.* 2015; Essabir *et al.* 2016), and epoxy and polyester resins (Durowaye *et al.* 2014; Ruggiero *et al.* 2016).

Research by Banat and Fares (2015) combined olive pomace and HDPE to make composites. The addition of a coupling agent improved the interfacial adhesion such that the olive pomace could act as a reinforcing material. Recycled HDPE (RHDPE) has been used in combination with banana fibers, a compatibilizer, and additional filler to generate composites. Banana fiber was a renewable, cost-effective, and non-abrasive filler used to increase the sustainability of an engineering thermoplastic (Satapathy and Kothapalli 2018). Other petro-based thermoplastics have been used as a matrix material. Polypropylene, for example, was combined with olive stone pomace by Naghmouchi *et al.* (2015) to develop composites. The researchers added maleic anhydride-grafted PP to improve the interfacial adhesion and further improve the mechanical performance (Naghmouchi *et al.* 2015). Coir fiber and shell particles were combined with PP by Essabir *et al.* (2016) to generate composites. Interestingly, the tensile modulus improved with the combination of the fiber and coir particles. The sustainability of the composites was improved, supporting their use with biobased and biodegradable polymers. Furthermore, the addition of coupling agents improved the performance of the fabricated materials (Essabir *et al.* 2016).

Unsaturated polyester resins (as a matrix material), a catalyst and an accelerator were combined with coconut shell fibers and palm fruit to generate composites. The optimal filler contents for coconut shell and palm fruit filler were 10 wt% and 20 wt%, respectively (Durowaye *et al.* 2014). Date stones, also known as the seeds, were used as the filler in resins to make composites *via* casting methods (Ruggiero *et al.* 2016). The addition of a low-cost filler helps to generate renewable materials for a more sustainable future. The reduction in cost, compared to neat polymer, is also an incentive for use in industrial production of composites. Aht-Ong and Charoenkongthum (2002) combined low-density polyethylene (LDPE), banana starch, and ethylene vinyl acetate copolymer (EVA). The EVA acted as a compatibilizing agent to improve the interactions between the filler and the matrix. These films functioned well, but there could be improvement with the sustainable content overall.

There has been limited research performed with tomato pomace and biobased or biodegradable plastics to date. However, post-harvest tomatoes have been used in combination with ethylene vinyl alcohol (EVOH) to make films (Nisticò *et al.* 2017). The films reduced the cost and overall consumption of petro-based plastic materials because tomato waste was able to act effectively as an additive. The addition of the natural filler improved the overall sustainability of the materials (Nisticò *et al.* 2017).

There is a popular misconception that the addition of natural fibers changes the biodegradability of non-biodegradable polymers when used to make composites. However, this is not the case. The addition of natural fillers or fibers increases the sustainability of the samples by increasing the renewable content but does not change the inherent nature of the continuous phase. If the continuous phase is not biodegradable, the addition of natural fibers will not change its biodegradability. Therefore, it is important to note that not all composites containing plant-based material are biodegradable, since their biodegradability is most determined by the matrix material.

Overall, this paper provides a comprehensive review of biodegradable composites containing natural fillers, highlighting the feasibility and importance of natural, renewable, and sustainable materials. Going forward, producers of plastics (especially single-use plastics) should keep in mind the end-of-life plan during product development to ensure a cleaner future for the environment. This ideology is discussed extensively by the Ellen MacArthur Foundation, calling it the “circular economy approach.” The circular economy approach, as shown in Fig. 1, illustrates how materials can be reused and recycled. In this work, the valorization of fruit waste re-uses an existing by-product to generate value-added and novel products (Ellen MacArthur Foundation 2017).

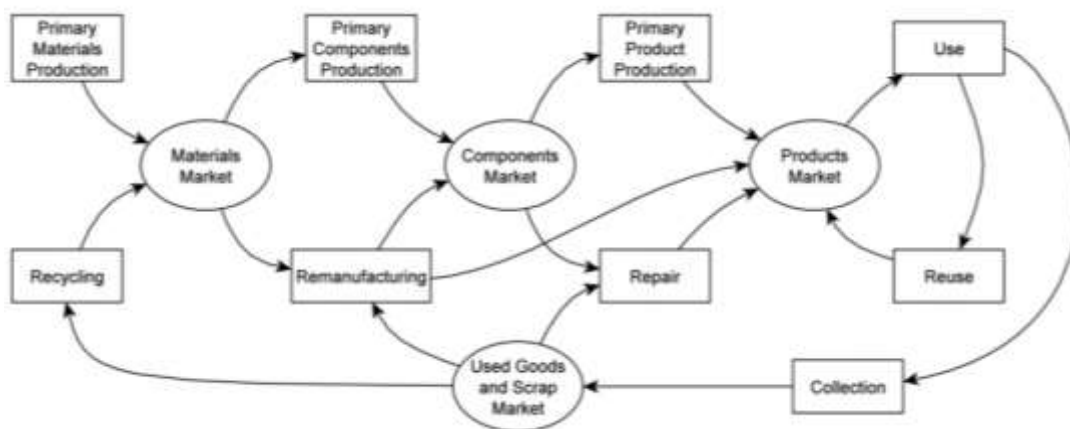


Fig. 1. A circular economy approach to sustainable product development. Reprinted with permission from John Wiley & Sons: *Journal of Industrial Ecology*, (Zink and Geyer 2017), Copyright 2017, License Number: 4561921510933.

BIODEGRADABLE POLYMERS

Biodegradable polymers can be degraded into water and carbon dioxide in the presence of microorganisms. Their degradation is strongly dependent on their surrounding environmental conditions, such as temperature and humidity (Mohanty *et al.* 2005). A polymer is deemed biodegradable when the polymer degrades during its application or soon after its application (Göpferich 1996). When a significant loss in properties occurs due to thermal, mechanical, or chemical degradation, or from photoexposure, a polymer can be deemed degradable (Göpferich 1996).

Degradable materials can be further specified as compostable. According to standards such as ASTM D5988-18 (2018) or ASTM D5338-15 (2015), a compostable plastic is a plastic that yields carbon dioxide, water, inorganic compounds, and biomass at a rate comparable to other known compostable materials and leaves no visible, distinguishable, or toxic residue. After 12 weeks there should be 10% of the original dry weight or less remaining when sieved on a 2 mm sieve (ASTM D5338-15 2015; ASTM D5988-18 2018).

Biobased or Renewable Resource-based Biodegradable Polymers

Biobased or renewable resource-based biodegradable polymers are polymers produced from renewable resources. Some renewable resources used to fabricate

polymers are plant-based, such as sugarcane or corn. Other renewable sources include bacterial fermentation of sugars. The most common biobased and biodegradable polymers include polylactic acid (PLA), polyhydroxyalkanoates (PHAs), and starch.

Polylactic acid (PLA)

Polylactic acid is a widely used, fully biobased polymer made from cassava, sugarcane, or corn. Polylactic acid is widely used due to its biodegradability, good mechanical properties, and comparatively low cost. There are different types of PLA, depending on the orientation of its monomer units. Lactic acid has two chiral carbons, which allow it to have different stereoisomers, namely L-lactide, D-lactide, and L-D-lactide. The properties of PLA are highly dependent on the stereochemical composition. When lactic acid (the precursor to PLA) is derived from biological sources, the majority of it is L-lactic acid, with a small amount being D-lactic acid. Therefore, PLA produced from biological sources is mainly composed of poly(L-lactide) (Garlotta 2001).

Polylactic acid is also a versatile material which can be processed in many ways, such as injection molding, thermoforming, blow molding, extrusion blown film, foaming, sheet extrusion, and film extrusion (Auras *et al.* 2010). The main disadvantages associated with PLA are its poor impact strength and low heat distortion temperature. Due to these disadvantages, many studies have been performed to overcome these properties by melt blending PLA with other polymers or additives to improve its properties (Mekonnen *et al.* 2013; Nagarajan *et al.* 2016).

Polyhydroxyalkanoates (PHAs)

Polyhydroxyalkanoates are polyesters created from bacterial fermentation. They can be synthesized by different strains of bacteria with each synthesized material exhibiting unique properties. The composition of the derived PHA polymer depends on the growth medium used (Reddy *et al.* 2003).

Polyhydroxyalkanoates can be classified into different groups, depending on their carbon numbers. Short chain length (SCL) PHAs have monomers with 2 carbon atoms to 5 carbon atoms, whereas medium chain length (MCL) polymers have monomers with 6 carbon atoms to 14 carbon atoms (Park *et al.* 2012). While there are many types of PHAs that can be synthesized, most are not commercially available. The most common industrially produced PHAs can be listed as medium chain length PHA (MCL PHA), polyhydroxybutyrate (PHB), and poly(hydroxybutyrate-co-valerate) (PHBV) (Chen 2009).

Polyhydroxybutyrate has poor melt processability and thermal stability, crystallizes slowly, and is brittle (Muthuraj *et al.* 2018). The properties of PHB can be modified and improved by copolymerization with hydroxyvalerate (HV), resulting in a product called PHBV, which, with its high HV content, has better ductility and toughness compared to PHB (Avella *et al.* 2000). Although PHBV is biocompatible, it has poor thermal stability and is expensive, difficult to process, and brittle. The crystallinity and polymer type of PHAs determine their degradation rate. The main uses for PHAs are plastic shopping bags, medical applications (such as tissue scaffolding and bone healing), agricultural films, fishing nets, and cosmetic packaging (Philip *et al.* 2007). Other researchers have reviewed many different articles on the preparation, properties, potential applications, and future development of PHAs (Avella *et al.* 2000; Philip *et al.* 2007).

Starch

Starch is a carbohydrate polymer that consists of many glucose units joined together by glycosidic bonds. It contains approximately 20% to 25% linear amylose and 75% to 80% helical/branched amylopectin by weight. It is found in many plants, such as potatoes, cereal grains, rice, maize, and cassava, among others. The characteristics of starches can differ depending on the amylose-to-amylopectin ratio, crystallinity, and glass transition temperature (T_g). The properties associated with starch, such as the high T_g (approximately 240 °C), strong inter- and intra-molecular hydrogen bonding, water sensitivity, and poor flowability of the starch granules, can limit processability and therefore reduce the amount of applications starch can be used in (Mekonnen *et al.* 2013).

These challenges can be overcome by the incorporation of plasticizers, blending with polymers, chemical modification, or a combination of these methods (Mekonnen *et al.* 2013). Starch can be plasticized with water, glycerol, glycol, sorbitol, fructose, mannose, fatty acids, *etc.* (Rosa *et al.* 2009; Luchese *et al.* 2018). Chemical modifications such as hydroxylation, acylation, oxidation, and acetylation have been reported to improve its processability and mechanical properties (Zamudio-Flores *et al.* 2010; Mekonnen *et al.* 2013). Although chemical modification is an effective way to modify starch, it can produce harmful by-products and is normally quite expensive. Therefore, plasticization is thought to be the best method to enhance the properties of starch.

Biobased and Organic Additives to Biodegradable Polymers

Biobased additives are derived from natural sources and are added to composites or films to improve their properties or material stability. Most often, the additives are used as plasticizers or binding agents.

Glycerol

Glycerol, also known as 1,2,3-propanetriol, is a simple polyol containing three hydroxyl groups. Animals, plants, and microorganisms naturally synthesize glycerol. Industrially, it can be produced from biobased or petro-based sources (Pagliaro *et al.* 2007). Here, the synthesis of biobased glycerol is the focus. Biobased glycerol can be obtained from saponification or from the hydrolysis of fats and oils. Saponification functions by creating soap from the fermentation of sugars to glycerol by microorganisms. It can also be obtained from transesterification of fats and oils in biodiesel production as a coproduct (Pagliaro *et al.* 2007). Glycerol is often used as a plasticizing agent when combined with other materials such as starch (Park *et al.* 2009; Deng and Zhao 2011).

Sorbitol

Sorbitol is a natural alcohol found in fruits and is a common plasticizer in biobased/edible films to aid extensibility and permeability (Shaw *et al.* 2002). It is the high molecular weight molecules that maintain a solid state at room temperature. When combined with film materials, it can impinge on the molecular interactions between molecules of the polymer, thereby reducing the brittleness and increasing flexibility of the films (Shaw *et al.* 2002). More specifically, sorbitol acts to increase the intermolecular spacing between molecules by reducing the hydrogen bonding between internal structures (McHugh and Krochta 1994).

Hexamethylenetetramine

Hexamethylenetetramine (HMTA) is a water-soluble organic compound made *via* a condensation reaction of organic compounds (Blažević *et al.* 1979). It is added to films such as polyvinyl alcohol (PVOH) to act as a cross-linking agent (Ooi *et al.* 2011; Zhong *et al.* 2011). Hexamethylenetetramine has also been used in other applications such as medicine.

Cellulose

Cellulose is one of the world's most abundant natural polymers. It consists of linear chain consisting of anhydro-glucose monomers attached through 1-4 β -linkages (Nazir *et al.* 2013). In some cases, cellulose can be extracted from fruits such as durian (Penjumras *et al.* 2016) and oil palm empty fruit bunches (Nazir *et al.* 2013). Cellulose can be used in many ways in composites applications, such as being used as a reinforcing agent in powder form, nanofibers, and nanocrystals (Penjumras *et al.* 2016; Gouw *et al.* 2017).

Petroleum-based Biodegradable Polymers

Some polymers that can be created either entirely or partially from petroleum-based feedstocks can also be biodegradable. Examples of these polymers are poly(butylene adipate-co-terephthalate) (PBAT), PVOH, and poly(butylene succinate) (PBS).

Poly(butylene adipate-co-terephthalate)

Poly(butylene adipate-co-terephthalate) is synthesized from 1,4-butanediol, adipic acid, and terephthalic acid *via* polycondensation reactions. It has good mechanical properties, which are comparable to the properties of LDPE, though its barrier properties are slightly different from those of LDPE (Costa *et al.* 2015). Poly(butylene adipate-co-terephthalate) can be processed with normal polymer processing techniques, including mixing, extrusion, and injection molding. The downside to utilizing PBAT is its relatively low degradation temperature (140 °C to 230 °C), which can cause it to degrade during processing (Costa *et al.* 2015).

Polyvinyl alcohol

PVOH is synthesized from poly(vinyl acetate) by full or partial hydroxylation. The amount of hydroxylation determines the characteristics of the material, such as its chemical properties, physical characteristics, and mechanical properties (Baker *et al.* 2012). PVOH is often used in medical applications as contact lenses, artificial cartilage, and meniscuses due to its biocompatibility, chemical resistance, high water solubility, and adsorption characteristics (Baker *et al.* 2012).

Poly(butylene succinate)

Poly(butylene succinate) is a biodegradable polymer that can be synthesized entirely from petroleum or be partially biobased, utilizing biobased succinic acid. It is created from 1,4-butanediol and succinic acid. Its properties are similar to PP and polyethylene (PE) (Fujimaki 1998). Poly(butylene succinate) can be processed easily by injection molding, sheet extrusion, thermoforming, blow molding, and compression molding.

Polycaprolactone

Polycaprolactone (PCL) is a semi-crystalline biodegradable polymer with a low melting temperature of 60 °C and a glass transition temperature of -60 °C. It is synthesized from ϵ -caprolactone *via* a ring-opening polymerization reaction. The degradation of the polymer is based on hydrolysis of the ester linkages. It has gained increasing interest in biomedical applications such as implants and drug delivery systems (McKeen 2012), as well as in sustainable packaging (Ahmad *et al.* 2018).

BIOFILLERS AND FIBERS

Biobased fillers and fibers have been widely used since the 1940s (Mohanty *et al.* 2000). Biofillers and fibers are advantageous because of their low cost, light weight, and abundance. Incorporating them into a composite increases the amount of biocontent in the composite, which in turn creates a more environmentally friendly product. The use of biofillers also reduces carbon footprints and improves energy security (Mohanty *et al.* 2018). Biofillers and fibers have other advantages such as reduced tool wear, good thermal properties, reduced worker respiratory irritation, and biodegradability (Mohanty *et al.* 2000). The downside associated with fillers or fibers is their hydrophilicity, which decreases compatibility with typical polymer matrices that are relatively hydrophobic. The other disadvantage is the low processing temperature to prevent degradation. On average, the thermal degradation temperature for fillers is approximately 180 °C, which limits their use in engineering thermoplastics. Some of these challenges are overcome by modifications, as discussed later.

















Fillers

Biofillers reinforce the strength and stiffness of a material. Their properties depend on their source, including what part of the plant they are taken from, the quality of the plant, and its age, among other factors. In this paper, all the fillers discussed are from fruits.

Fillers are typically composed of cellulose, hemicellulose, lignin, and protein because they are not limited to only the fibrous component of plants. Variations in growing season and preparation of the filler can influence its properties, along with the components included in the filler.

Natural fillers considered in this review include apple, banana, acai berry, blueberry, cranberry, coconut, grape, durian, rambutan, olive, mango, pineapple, and date (Table 1). In many cases, the waste is a combination of the skin, flesh components, and seed components. However, some stem materials and leaves would be present and could be processed from these biomass sources.

Table 1. Fruits Used as Filler or Fiber in Composites and Films

<p>Apple</p> 	<p>Banana</p> 	<p>Blueberry</p> 	<p>Coconut</p> 
<p>Cranberry</p> 	<p>Acai Berry</p> 	<p>Durian</p> 	<p>Grape</p> 
<p>Mango</p> 	<p>Olive</p> 	<p>Pineapple</p> 	<p>Rambutan</p> 
<p>Date and Palm Fruit Bunch</p> 	<p>Tomato</p> 	<p>Kiwi Skin</p> 	<p>Jackfruit</p> 

Quantity of Fillers

The global production of the various fruits is shown in Table 2. The average waste generated from whole fruits is approximately 30% (Vendruscolo *et al.* 2008). Wastes generated from apples, grapes, and olives are commonly referred to as “pomace” or “marc,” as they are pressed to extract the liquid components for applications such as making juice or wine.

Table 2. Global Fruit Production in Weight of Whole Fruit

Fruit	Global Production	Waste (%)	Total Waste
Apple	~74.2 million tonnes (2018) (USDA 2018)	20 (Gouw <i>et al.</i> 2017)	14.8 million tonnes
Grape	~23.4 million tonnes (2018) (USDA 2018)	25 (Dwyer <i>et al.</i> 2014)	5.85 million tonnes
Banana	~114 million tonnes (2017) (FAO 2019a)	30	34.2 million tonnes
Pineapple	~25.8 million tonnes (2017) (Altendorf 2019)	30	7.74 million tonnes
Mango	~48.3 million tonnes (2017) (Altendorf 2019)	30	14.49 million tonnes
Blueberry	~0.59 million tonnes (2017) (FAO 2019b)	30	177 thousand tonnes
Coconut	~60.8 million tonnes (2017) (FAO 2019b)	30	18.24 million tonnes
Coir	~1.23 million tonnes (2017) (FAO 2019b)	30	369 thousand tonnes
Date	~8.17 million tonnes (2017) (FAO 2019b)	30	2.45 million tonnes
Kiwi	~4.04 million tonnes (2017) (FAO 2019b)	30	1.21 million tonnes
Olive	~20.9 million tonnes (2017) (FAO 2019b)	35 (Banat and Fares 2015)	7.32 million tonnes
Cranberry	~0.63 million tonnes (2017) (FAO 2019b)	30	189 thousand tonnes
Durian	~3 800 tonnes (2016) (Lee <i>et al.</i> 2018)	60 (Lee <i>et al.</i> 2018)	2 280 tonnes

The pomace materials are usually abundantly available but serve very little purpose. In fact, Gouw *et al.* (2017) estimated that only 20% of apple pomace generated is used for a value-added purpose, whereas the rest remains animal feedstock or is left for compost. Based on the global production of fruit, there are substantial amounts of biomass waiting to be repurposed.

The total waste was calculated by multiplying the global production by the estimated weight percentage of waste, *i.e.*, approximately 30% for fruit. Based on the estimated total wastes generated from fruit processing, there are apparently millions of tonnes of materials in need of a value-added purpose. Currently, some of the waste is used in biorefining and energy production, chemical extraction to generate useful products, or animal feedstocks. However, fruit waste that ends up in landfills or as compost does not possess any sort of added benefit but, rather, generates environmental burden. Therefore, if the biomass is instead used to produce value-added products, then the carbon is sequestered, the impact on the environment is reduced, and the overall sustainability of the materials is improved.

Physicochemical composition of fillers

As mentioned earlier, cellulose, hemicellulose, and lignin are among the major constituents of biobased fillers. Each fruit possesses a unique combination of each of these fractions (Table 3), affected by the agricultural conditions (Muensri *et al.* 2011). Cellulose is often located within the backbone structure of the fiber. In contrast, lignin is a polyphenolic compound of amorphous structure, located on the outside of the fiber

(Muensri *et al.* 2011). The lignocellulosic materials in the natural fiber are responsible for cross-linking to the polymer matrix. More cross-linking between the filler and the matrix results in better interfacial adhesion, which enhances the filler's ability to distribute stresses throughout the matrix materials. Better interfacial adhesion often results in improved impact strength of composites (Picard *et al.* 2019). The lignocellulose content can also affect other properties of the composites.

Table 3. Cellulose, Hemicellulose, and Lignin Contents of Various Fruit Biomasses

Fruit	Cellulose (%)	Hemicellulose (%)	Lignin (%)	Reference
Apple	47.49	27.77	24.72	Guerrero <i>et al.</i> (2014)
Grape	17.5	6.9	51.7	Gómez-Brandón <i>et al.</i> (2011)
Coconut husk	34	21	27	Bledzki <i>et al.</i> (2010)
Pineapple leaf	66.2	19.5	4.28	Daud <i>et al.</i> (2014)
Banana fiber	63-64	19	5	Deepa <i>et al.</i> (2011)
Durian	60.45	13.09	15.45	Aimi <i>et al.</i> (2014)
Coir fiber	32.69	22.56	42.10	Muensri <i>et al.</i> (2011)
Olive stone flour	25	34	34.5	Naghmouchi <i>et al.</i> (2015)
Mango peel	9.19	14.51	4.25	Sánchez Orozco <i>et al.</i> (2014)
Post-harvest tomato	33.1	10.5	7.8	Nisticò <i>et al.</i> (2017)
Rambutan peel	24.28	11.62	35.34	Oliveira <i>et al.</i> (2016)
Palm oil empty fruit bunch	37.26	14.62	31.68	Sudiyani <i>et al.</i> (2013)

Thermal stability of composites is strongly dependent on the presence of natural filler. Thermogravimetric analysis reveals that hemicellulose is the first lignocellulosic material to degrade between 150 °C and 350 °C (Yang *et al.* 2005). Cellulose and lignin are the next to degrade, at 275 °C to 350 °C and 250 °C to 500 °C, respectively (Kim *et al.* 2005). It can be inferred that samples with greater hemicellulose contents, relative to cellulose and lignin, would be less thermally stable and therefore be recommended for combining with plastics that have lower melting points.

Fibers

In composite applications, fibers enhance the strength and stiffness of a material. The properties of the fibers depend on their source, *i.e.*, whether they are from the stem of the plant or the leaves of the plant, the quality of the plant, and the age of the plant. Natural fibers can be placed into different groups: leaf, bast, seed, and fruit. This review will focus on fruit leaf and fruit fibers, as they are both by-products of food processing. Natural fibers exhibit variation in length and diameter. Other factors determining the properties are size, maturity, and the processing method used for extraction of the fibers (Mohanty *et al.* 2000).

Modification of Fibers or Filler

Surface modification of fibers is needed for improving their performance. In composite applications, strong fiber-matrix interaction is crucial for good mechanical properties (Mohanty *et al.* 2018). However, natural fibers are hydrophilic, which presents a problem when they are added to a polymer matrix that is hydrophobic. Fibers also tend to have a waxy coating on their surface, which causes weak matrix bonding and poor

surface wetting (Mohanty *et al.* 2001). Therefore, surface modification to reduce the hydrophilicity of the fibers can improve the matrix adhesion. Modifications such as washing, peroxide treatment, alkali treatment, bleaching, and the use of silane coupling agents have yielded fiber enhancement (Mohanty *et al.* 2001). Not only does surface treatment improve the interfacial adhesion, but it can also aid in filler dispersion (La Mantia and Morreale 2011).

Each of the different modification methods has advantages and disadvantages. For example, silane treatments usually result in strong enhancement of mechanical properties. For thermal stability, alkali treatments and acetylation are usually more favorable (La Mantia and Morreale 2011). However, chemical modification is costly and complex, thereby limiting its industrial relevance. Due to these limitations, the most common approach to enhancing natural filler/fiber composites is to add small amounts of compatibilizer to enhance the material properties. The compatibilizer should have both hydrophilic and hydrophobic moieties, which interact with polar groups on the filler's surfaces and polymer molecular chains, respectively. Although the addition of compatibilizer is common, it is not the focus of this review.

Washing

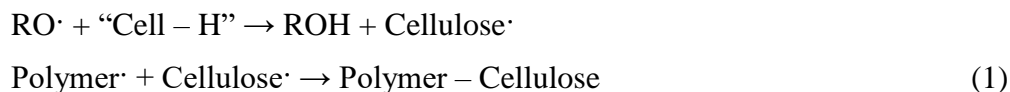
Washing of fibers is a routine step when modifying fibers. Washing fibers in water alone or with water and a mild detergent removes dust or any other impurities that could be present. Fibers are usually also washed after chemical modification to remove any residual chemicals. Washing has also been used to remove excess free sugars in some biomass sources to improve the thermal stability at polymer processing temperatures.

Bleaching

Bleaching of fibers is done to remove colour and/or to modify the surface properties. In lignocellulosic fibers, the colour of the fibers can be associated with lignin (Razak *et al.* 2014). Bleaching also enhances mechanical properties, as it can increase the surface roughness of the fibers, resulting in stronger fiber-matrix adhesion (Razak *et al.* 2014).

Peroxide treatment

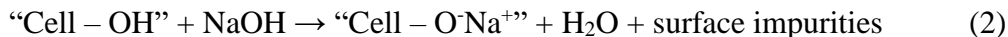
Peroxides are molecules with an O-O group. Peroxides can decompose easily, forming free radicals in the form of RO·, which then react with the hydrogen groups on the surface of the fibers. For example, the hydrogen on the cellulose (Cell-H) interacts with RO·, which in turn can react with the polymer as shown in Eq. 1 (Joseph *et al.* 1996):



Peroxide treatment can remove surface impurities, hemicellulose, and lignin and can also increase the surface roughness of the fibers, increasing the fiber-matrix interaction (Razak *et al.* 2014). For peroxide treatments, fillers/fibers are added to a peroxide solution and left to soak. The fillers/fibers are washed to remove any residual peroxide and then dried.

Alkali treatment

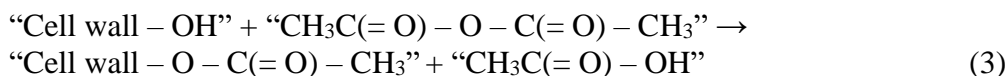
Alkali treatment, also called “mercerization” if the concentration is high enough, is a low-cost and effective method for surface modification of fibers. The reaction between sodium hydroxide (NaOH) and cellulose fiber (Cell-OH) is shown in Eq. 2 (Mohanty *et al.* 2001):



This treatment is effective in removing lignin, wax, and oil that cover the external surface of the fiber, depolymerizing the cellulose structure, and exposing crystallites of short length (Mohanty *et al.* 2001). It also increases the surface roughness by disrupting the hydrogen bonding in the network structure (Li *et al.* 2007). During alkali treatment, fibers are immersed in NaOH solution. The fibers are then removed and dried. The NaOH concentration and soaking time need to be optimized because high NaOH concentration or extended soak duration can result in undesired fiber characteristics (Li *et al.* 2007). Typically, low concentrations of NaOH (approximately 5% or less) are incorporated with water in a bath. The filler/fibers are then soaked for a short duration, from 30 min to a few hours. The filler/fibers are then washed to remove the NaOH and dried to remove moisture.

Acetylation treatment

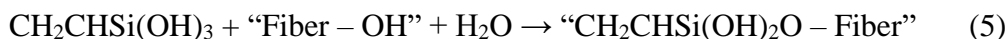
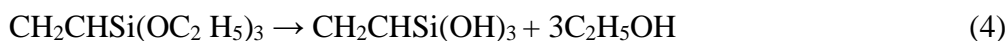
Acetylation is a type of esterification method that introduces an acetyl group into a compound. The acetic anhydride addition to the lignocellulosic components of the biofiller/fiber causes an esterification reaction with the hydroxyl groups in the cell walls of the biofiller/biofiber (Rowell 2004). Acetic acid is formed as a by-product of the reaction. This reaction is shown in Eq. 3 (Rowell 2004):



For acetylation, biofillers/fibers are soaked in water and then filtered and placed in an acetylation solution. The temperature, duration, and exact chemicals used can vary, but the fillers/fibers are typically washed and dried after this process (Bledzki *et al.* 2008)

Silane treatment

Silane is an inorganic compound with the chemical formula SiH₄. Silane may reduce the amount of cellulose hydroxyl groups on the surface of the filler and reduces the fiber-matrix interaction (Li *et al.* 2007). Silane reacts with water and forms silanol and alcohol. The silanol then reacts with the OH groups on the fiber, resulting in stable covalent bonds on the surface of the fiber (Agrawal *et al.* 2000). The proposed reaction is shown in Eqs. 4 and 5 (Agrawal *et al.* 2000):



The swelling of the fiber is prevented because the silane creates a crosslinked network of covalent bonds between the fiber and matrix (Li *et al.* 2007). There are many different silanes used, along with different concentrations, such as 1 M NaOH for 1 h at room temperature (Jandas *et al.* 2011). For example, long-chain alkyl methoxysilanes can be used to prepare hydrophobic plant-based fibers (Sgriecia *et al.* 2008). For silane treatments, the fillers/fibers are soaked in a mixture containing a small amount of silane,

with a mixture of water and ethanol or a solvent mixture such as toluene/ethanol/acetone (Shih *et al.* 2014; Hemsri *et al.* 2012). After soaking for a specific duration, the fibers are washed and dried. The specific conditions depend on the silane used and the desired properties.

Plasma treatment

Plasma treatment is a rather recent development for changing the surface properties of natural fillers/fibers without significantly affecting their bulk properties (Kalia *et al.* 2009). A plasma treatment is based on allowing ionized gas with an equal number of positively and negatively charged molecules to react with the surface of the fillers/fibers (Kalia *et al.* 2009). The treatment conditions used heavily influence the final properties of the treated material, so it is difficult to generalize the final properties (La Mantia and Morreale 2011). Podgorski and Roux (1999) and Podgorski *et al.* (2000) determined that the type of gas, the treatment duration, the power, and the distance between the samples and the plasma source all influenced the properties of the plasma-treated biofillers/fibers.

PROCESSING METHODS

Processing methods for the production of composites with plant-based fillers include injection molding, compression molding, and compounding (Rout *et al.* 2001; Jandas *et al.* 2012; Dong *et al.* 2014; Koutsomitopoulou *et al.* 2014; Hassaini *et al.* 2017). The method chosen usually depends on how the incorporation of the biofiller/fibers is performed. Woven fibers are usually processed with compression molding, while short fibers are usually injection molded. The end application is also considered when determining which processing method to use. The processing method chosen also reflects the thermal stability of the added biofiller/fiber, as many naturally sourced materials have low thermal stability, limiting processing temperatures to less than approximately 200 °C.

Injection Molding

Injection molding is a popular processing method for plastics due to the wide variety of materials that can be used and its ease of use. Injection molding is often used in composite applications because it has been shown to improve the fiber dispersion and increase the tensile and flexural properties (Mohanty *et al.* 2004). Conversely, injection molding can reduce fiber lengths due to the high shear the fibers are exposed to during the extrusion and injection conditions, which in turn changes the properties of the composites.

Compression Molding

Compression molding is a widely used composite fabrication technique due to its low cost and simplicity. One of the major benefits of compression molding is the amount of control over fiber orientation. The fibers can be randomly orientated, creating a composite with isotropic properties, or they can be selectively orientated, as desired. Certain natural fibers can be difficult to disperse during compression molding.

Every different composite formulation will have a preferred processing method. This can be determined based on previous work with similar polymer matrices and biofiller/fiber type, or by trial and error. Processing techniques include creating layers of

a fine polymer powder between a biofiller/fiber, sandwiching long fibers between layers, sandwiching short fibers or fillers between layers, dry mixing of components, and multiple layers of both powdered polymer and fibers until the desired weight percentage of material is used (Akil *et al.* 2011).

Casting

Casting is a common method used to make films. Casting requires the preparation of a polymer solution, which is poured into a mold and dried under ambient conditions or oven-dried at an elevated temperature. Different materials are used in combination with water or other solvents to form the solution for film production. These materials include starch and PVOH. Starch is a low-cost, versatile, and readily available biodegradable material with excellent film-forming properties (Luchese *et al.* 2018). Polyvinyl alcohol, however, is a water-soluble polymer that is formed *via* polymerization and hydrolysis of vinyl alcohol. Polyvinyl alcohol is a biodegradable polymer with high film strength. Plasticizing agents, such as sorbitol and glycerol, are being used to impart flexibility to the final film structures (Luchese *et al.* 2018).

BIOCOMPOSITES AND FILMS

Biocomposites derived from both natural fillers/fibers and biodegradable plastics can be classified as biodegradable materials. Despite much research on natural fiber composites, studies related to incorporating food waste into biocomposites are limited. Most of the research related to biocomposites from food waste focuses on biomasses such as coconut, olive, pineapple, and banana. Composites derived from these biomasses and other fruits are discussed in detail, focusing on their mechanical and thermal properties. The properties of the biocomposites depend heavily on the type of polymer matrix used, the treatment of the fibers, the amount of fibers added, the potential addition of compatibilizer, and the process techniques, among other factors. In general, the addition of compatibilizer and surface treatment of the biofiller/fiber typically improved the mechanical properties of the biocomposites.

Fruit Wastes in Composites

Various fruit wastes, such as blueberry, cranberry, apple, acai berry, rambutan, kiwi, mango, palm fruit, date fruit, and jackfruit, have been used to generate biocomposites and films.

Park *et al.* (2009) created blueberry, cranberry, and grape pomace biocomposite boards (Fig. 2) with soy flour modified with NaOH. They found that the blueberry pomace had the greatest breaking strength and modulus of elasticity values, as compared to the other pomace biocomposites. The addition of glycerol, acting as a plasticizer, increased the flexibility and decreased the stiffness of the test specimens (Park *et al.* 2009). The degradation temperature shifted slightly lower with the addition of the blueberry pomace, which was attributed to the lower thermal stability of the biofillers.

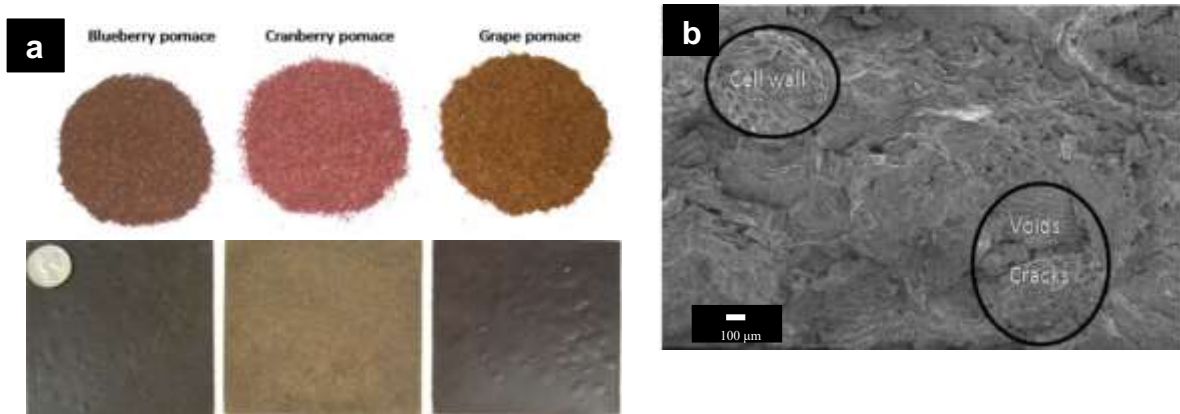


Fig. 2. (a) Blueberry, cranberry, and grape pomaces and their corresponding composites, (b) scanning electron microscope (SEM) image of fracture surface of blueberry pomace board. Reprinted with permission from John Wiley & Sons: *Journal of Applied Polymer Science*, (Park *et al.* 2009), License Number: 4561941149394.

Wataya *et al.* (2015) developed acai fiber-filled PBAT/PLA biocomposites. They found that 15 wt% of acai fiber decreased the tensile strength, Young's modulus, and impact strength. However, the elongation at break increased by approximately 17%. The melting temperature remained practically unchanged with the addition of fiber (from 178.0 °C to 178.5 °C) (Wataya *et al.* 2015). Apple pomace (AP) can be generated from bruised apples, jam production, and juice/cider production. This material is abundant around the world (Shalini and Gupta 2010). Gaikwad *et al.* (2016) created AP/PVOH films with antimicrobial properties. The AP/ PVOH film with 30 wt% AP had 40% scavenging activity on a free radical scavenging assay when using 2,2-diphenyl-1-picrylhydrazyl (DPPH), whereas the control with just a PVOH film had no scavenging activity. This result indicated that the AP films had a high antioxidant capacity. The authors attributed this result to the phenolic compounds within the AP (Gaikwad *et al.* 2016).

AP fiber was studied by Gowman *et al.* (2019) *via* scanning electron microscopy (SEM), showing porous and sheet-like morphologies (Fig. 3a). The different morphologies were attributed to the components of the pomace, including skin, seed, and flesh materials (Gowman *et al.* 2019). Picard *et al.* (2019) combined AP with biobased PBS (BioPBS) to generate biocomposites, with maleic anhydride-grafted BioPBS as a compatibilizer to improve the mechanical properties. The researchers found that the addition of AP improved the impact strength by more than 120%, even without the addition of compatibilizer. An SEM image of these composites is shown in Fig. 3b. The flexural strength and modulus of the composites with compatibilizer were greater than those of the neat BioPBS. Furthermore, SEM analysis determined that the addition of the compatibilizer improved the interfacial adhesion of the samples (Fig. 3c). The AP-based sustainable biocomposites may find single-use applications for food.

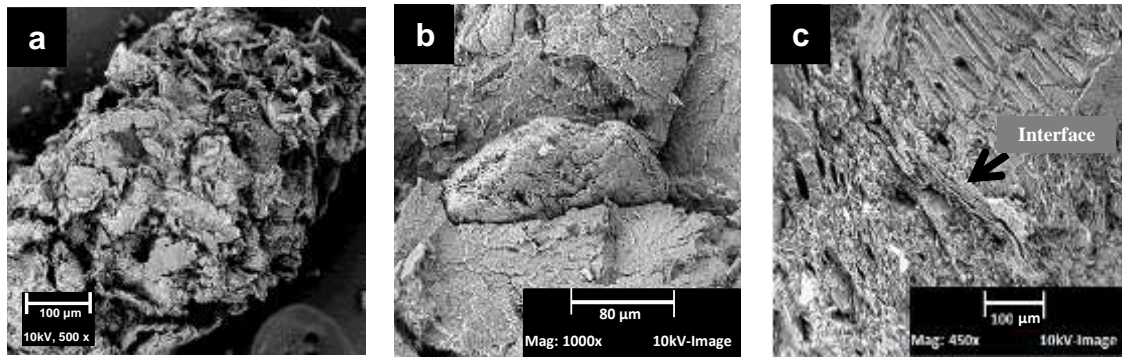


Fig. 3. (a) AP fiber (Gowman *et al.* 2019), (b) AP composites (Picard *et al.* 2019), (c) AP-compatible composites. Reprinted with permission from Springer Nature: *Waste and Biomass Valorization*, (Picard *et al.* 2019), License Number: 4560191136299.

Luchese *et al.* (2018) created blueberry pomace and cassava starch films. They found that the addition of blueberry pomace to their films caused high absorption of wavelengths less than 300 nm, meaning that the pomace was able to protect the films from UV light. The improved UV resistance was attributed to the presence of aromatic compounds in the blueberry pomace, which could help extend the shelf life of foods if used for food packaging applications.

Mittal *et al.* (2015) created date seed powder (DSP) and PLA/PBAT biocomposites. The authors found that 40 wt% of DSP increased the tensile modulus of their PBAT biocomposites by more than 300%, while with a PLA matrix, an increase was seen until reaching 20 wt% (Mittal *et al.* 2015). The melting point for the PBAT composites decreased with increasing DSP, but it remained almost the same for the PLA composites (Mittal *et al.* 2014). Voids were present and were explained by moisture evaporation during processing. The authors also noticed oil migration from the DSP but did not notice a change in the mechanical properties of the composites (Mittal *et al.* 2015). The biodegradation was improved with the addition of the DSP. Increased biocontent is known to improve degradation of composites, and in this case it resulted in bigger cracks and surface degradation in the samples (Mittal *et al.* 2014).

Rambutan (*Nephelium lappaceum*) is a tropical fruit that consists of an outer layer, flesh, and seed components. Cast films were prepared with PVOH, glycerol, HMTA, polysorbate 80 as a matrix material, and rambutan skin flour at approximately 8 wt% to 32 wt% filler content. Increasing rambutan flour content resulted in decreased tensile strength. Other properties such as water absorption were increased due to the hydrophilic nature of the filler. This can be less favourable for the use of the composite in food packaging or other applications. However, rambutan did show enhanced adhesion and stronger interaction with the matrix material than did banana skin flour in the same study (Zhong *et al.* 2011).

Durian (*Durio zibethinus*) is a commonly consumed fruit in Southeast Asian countries. It consists of 50% to 65% flesh components, and the remaining skin and seed materials are waste. Manshor *et al.* (2014) reported that NaOH-treated durian skin fibers possessed higher impact strength than untreated samples. However, there was little improvement to the overall impact strength of PLA. The authors concluded that the overall performance was comparable to PLA and suggested the use of this filler for cost-reduction (Manshor *et al.* 2014). In another study, durian husk fiber was combined with PLA at 15 parts per hundred rubber (phr), 30 phr, 45 phr, and 60 phr (Lee *et al.* 2018).

The mixtures were compression molded to generate tensile samples. Although tensile strength was lower in the composites as compared to the neat polymer, an increasing tensile strength was observed with increasing fiber content. Furthermore, the tensile modulus increased beyond that of the neat polymer as the fiber content increased. The authors highlighted that the samples were quite brittle (Lee *et al.* 2018). Thus, further work needs to be done to enhance the interactions of the fiber and matrix and further improve the mechanical properties of the composite.

Table 4. Fruit Pomace and Fruit Waste Composites

Fruit	Treatment	Matrix	Preparation Method	Reference
Acai fiber powder (15 wt%)	Washed	PBAT/PLA	Injection molding	Wataya <i>et al.</i> (2015)
Apple 1 wt%, 5 wt%, 10 wt%, 30 wt%	Untreated	PVOH	Films	Gaikwad <i>et al.</i> (2016)
Apple 20 wt%, 30 wt%, 40 wt%, 50 wt%	Washed	BioPBS	Injection molding	Picard <i>et al.</i> (2019)
Blueberry 20 wt%	Untreated	Soy flour or pectin/xanthan gum and glycerol	Injection molding	Park <i>et al.</i> (2009)
Blueberry 4 wt%, 8 wt%, 12 wt%	Bleached	Cassava starch and sorbitol	Films	Luchese <i>et al.</i> (2018)
Cranberry 20 wt%	Untreated	Soy flour or pectin/xanthan gum and glycerol	Injection molding	Park <i>et al.</i> (2009)
Date seed powder 10 wt%, 20 wt%, 30 wt%, 40 wt%	Cleaned with sulfuric acid	PLA or PBAT	Injection molding	Mittal <i>et al.</i> (2014, 2015)
Rambutan skin 8 wt% to 32 wt%	Untreated	PVOH, glycerol, HMTA, polysorbate 80	Film	Zhong <i>et al.</i> (2011)
Durian skin 0 wt% to 40 wt%	Untreated and treated with 4 wt % NaOH	PLA	Injection molding	Manshor <i>et al.</i> (2014)
Durian husk 15 phr, 30 phr, 45 phr, 60 phr	Untreated	PLA	Compression molded test bars	Lee <i>et al.</i> (2018)
Mango: lump and almond ~8 wt%	Treated: Sanitization	PLA	Films	Lima <i>et al.</i> (2019)
Kiwifruit skin	Unknown	PLA	Industrial production	Graichen <i>et al.</i> (2017)
Palm fruit bunch 12.2 wt% to 63.8 wt%	Washed	PCL	Compression molded test bars	Ahmad <i>et al.</i> (2018)
Jackfruit skin 8 wt% to 32 wt%	Untreated	PVOH, glycerol, polysorbate 80, HMTA	Films	Ooi <i>et al.</i> (2011)

Mango (*Mangifera indica* L.) is a tropical fruit. Considerable waste is generated as tegument from the outer skin and the seed. Lima *et al.* (2019) developed films combining PLA with mango seed fiber and/or the tegument. X-ray diffraction analysis (XRD) patterns of the composite displayed a shift in peaks due to the presence of starch

and cellulose from the samples. Interestingly, the presence of either the seed or tegument increased the tensile strength and Young's modulus over that of the neat PLA film. The increase in modulus was attributed to an anchoring surface generated between the cellulose fibers of the mango parts with PLA, which led to better stress transfer in the system (Lima *et al.* 2019). Similarly, kiwifruit skin has been combined with PLA to produce a bio-spife, which is a biodegradable utensil combining a spoon and a knife. The bio-spife is manufactured as an alternative to current market spifes generated from polystyrene. The bio-spife was generated with mechanical properties similar to that of spifes made from polystyrene, but it is compostable and hence more sustainable than its petroleum counterpart (Graichen *et al.* 2017).

Ahmad *et al.* (2018) combined oil palm empty fruit bunches (OPEFB) with PCL to make biodegradable composites. The thermal stability of the composites decreased, a common result when natural fibers are added to polymer as fillers. Fourier-transform infrared spectroscopy analysis revealed that the interactions between the fiber and matrix material were mainly physical (Ahmad *et al.* 2018). Overall, the addition of the fillers increased the biocontent and improved the sustainability of the biocomposites. Biodegradable films have also been developed from jackfruit (*Artocarpus heterophyllus*) waste and PVOH. Ooi *et al.* (2011) found a decrease in tensile strength with increasing filler loading. However, the Young's modulus increased with increasing jackfruit waste, suggesting that the filler acted as a stiffening agent (Ooi *et al.* 2011). All fruit pomaces discussed in this section are summarized in the Table 4.

Grape Pomace

Grape pomace is generated during juice or wine production. This biomass consists of the seeds, skins, flesh, and stems of the grapes. Notably, the chemical composition of the pomace materials would likely be different from study to study; moreover, samples could be fresh or partially fermented. Some winemaking processes require the grape pomace to remain in solution during the winemaking process. This results in the use of the sugars in the samples to fuel the fermentation process. Therefore, pomace generated from juice or wine without fermentation of the pomace contains more sugar. The sugar content of the pomace samples can affect their thermal properties.

Jiang *et al.* (2011) created both white and red grape pomace biocomposites with grape skin and soy flour or soy protein isolate/PVOH blend. They added stearic acid and epichlorohydrin at different weight percentages to optimize the material properties. Both the white and red grape pomace biocomposites were optimized in terms of breaking strength and modulus of elasticity, water sensitivity, thermal degradation temperature, and soil biodegradation (Jiang *et al.* 2011). The weight losses for the biocomposites after 30 d in soil were 50% and 80% for the red and white grape skin boards, respectively. The red and white grape skin biocomposites had different properties and optimized formulas due to their different chemical compositions and the processing methods used (Jiang *et al.* 2011).

Spiridon *et al.* (2016) created grape seed and PLA biocomposites with 3 wt% and 10 wt% grape seeds. The incorporation of grape seeds reduced thermal stability but improved the stiffness of the matrix. The addition of the grape seeds also improved the accelerated aging characteristics of the biocomposites, when compared to neat PLA (Spiridon *et al.* 2016). The grape seed biocomposites had antimicrobial activity toward Gram-positive and Gram-negative bacteria, which could be advantageous in food packaging applications (Spiridon *et al.* 2015). Similarly, Gowman *et al.* (2018) developed

grape pomace (Fig. 4a) and BioPBS-based biocomposites (Fig. 4b and 4c). They found that 40 wt% GP with 3 wt% maleic anhydride-grafted BioPBS (MA-g BioPBS) had the best balance of mechanical and thermal properties. This biocomposite formulation showed 28.4% and 59% improvements in flexural and impact strengths, respectively, while the heat distortion temperature increased by 14.3% (Gowman *et al.* 2018).

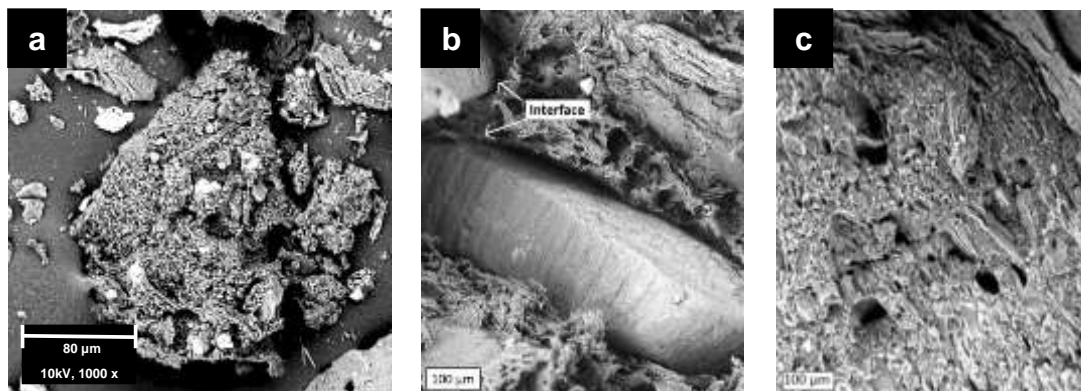


Fig. 4. (a) Grape pomace fiber (b) grape fiber in composites, (c) compatibilized grape pomace composites (Gowman *et al.* 2019).

Table 5. Grape Filler-based Composites and Films

Fruit	Treatment	Matrix	Preparation Method	Reference
Red grape skins and white grape skins	Untreated	Soy flour, PVOH, soy protein isolate, stearic acid, and epichlorohydrin	Injection molding	Jiang <i>et al.</i> (2011)
Grape 3 wt% and 10 wt%	Untreated	PLA	Injection molding	Spiridon <i>et al.</i> (2015, 2016)
Grape 20 wt%, 25 wt%, 30 wt%, 40 wt%, 50 wt%	Untreated	PBS and MA-g BioPBS	Injection molding	Gowman <i>et al.</i> (2018)
Grape 98 wt% to 99 wt%	Treated	Polysaccharide film mixture, LMP, glycerol	Film	Deng and Zhao (2011)

Deng and Zhao (2011) combined fresh red grape pomace with polysaccharides to generate biobased food-grade films. The main constituents in the films were a polysaccharides mixture (of sodium alginate, carrageenan, cell and cellulose gum), and low-methoxyl pectin (LMP). Glycerol was added to some blends to act as plasticizer. Overall, pomace was an effective colouring agent in the films, in addition to acting as a water barrier. Meanwhile, mechanical properties of the biocomposites were satisfactory. Furthermore, the release of phenolic compounds during the film-making process suggested the potential for the film to possess antimicrobial and antioxidant properties with further work (Deng and Zhao 2011). Overall, there have been many studies which incorporate grape pomace into films or composites (Table 5).

Olive

Olive (*Olea europaea*) is a fruit grown mostly for oil but also for its wood, leaves, or fruit. Olive pomace, or oil solid waste, is a mixture of skin, pulp, and pit, which are leftovers after pressing the olive for oil. Olive pomace can be re-pressed to produce olive

pomace oil, with the pomace still remaining as a by-product. Approximately 30% to 35% of the weight of the olive is pomace, generating large amounts of low-value by-product (Banat and Fares 2015).

For incorporation into composites, the olive by-products are dried and ground. The filler is washed to remove any impurities, although this is not always done. Most of the biocomposites containing olive filler involve a PLA matrix (Figs. 5a and 5b). Components of the olive incorporated include pit, husk, and pomace/waste/cake.

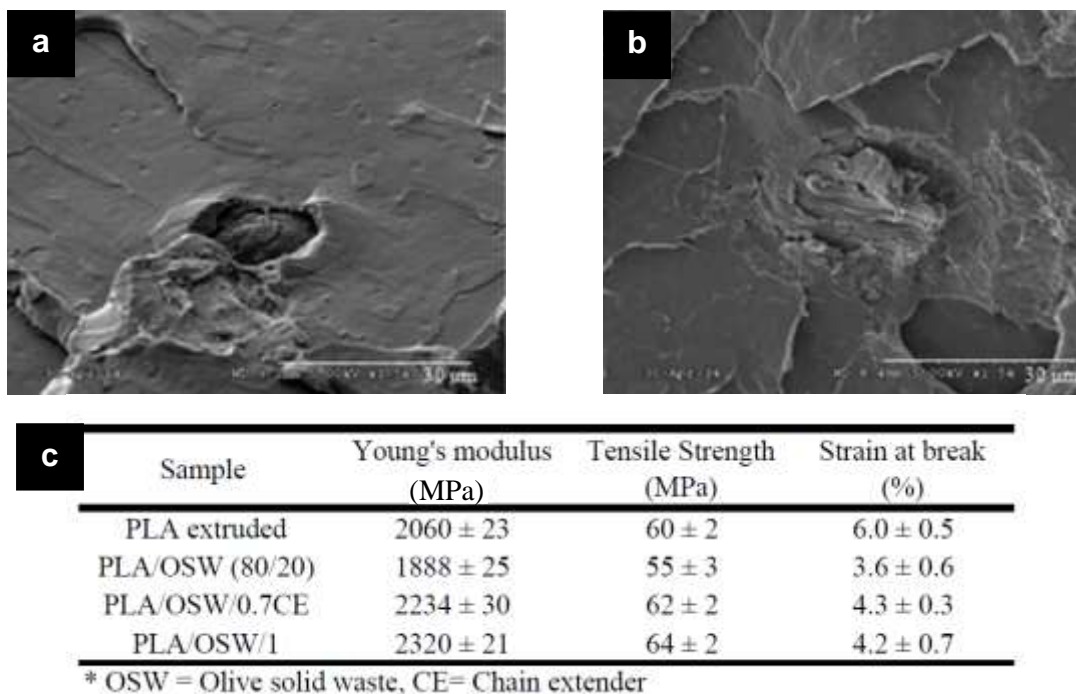


Fig. 5. (a) Olive waste and PLA, (b) olive waste and PLA with chain extender, (c) mechanical properties. Reprinted with permission from John Wiley & Sons: *Polymer Composites*, (Khemakhem *et al.* 2018b), Copyright 2016, License Number: 4557810973384.

Koutsomitopoulou *et al.* (2014) created biocomposites with olive pits and PLA. They investigated the effects of particle size and weight fractions on the mechanical and physical properties. An increase in tensile modulus and a small decrease in tensile strength were reported with the addition of 20 wt% olive pit powder (Koutsomitopoulou *et al.* 2014). Olive husk biocomposites have been made with various matrix materials, including poly(butylene succinate-co-butylene adipate) (PBSA) (Tserki *et al.* 2006), PLA (Isadounene *et al.* 2017), and PHBV (Hassaini *et al.* 2017). Tserki *et al.* (2005) treated the olive husk with acetylation and propionylation processes to esterify the olive husk and decrease its hydrophobicity. They found that the incorporation of 30 wt% olive filler increased the tensile modulus, water absorption, and the degradation rate. However, tensile strength was reduced. They also found that the treated olive had half as much water absorption as compared to the untreated olive (Tserki *et al.* 2006). Isadounene *et al.* (2017) showed that untreated olive husk with PLA increased the Young's modulus and that the incorporation of olive filler increased the storage modulus but decreased the elongation at break and the tensile strength (Isadounene *et al.* 2017). The authors then tested the effect of particle treatment on accelerated aging. The olive husk underwent an alkali treatment in NaOH and had improved mechanical properties but a lower

degradation rate, as compared to its untreated counterparts (Isadounene *et al.* 2018). Hassaini *et al.* (2017) investigated the effect of silane-treated olive husk on water contact angle, crystallinity, and tensile properties and compared with untreated olive husk. The addition of 20 wt% olive husk caused the crystallinity of PHBV to increase by 33.9% and 14.7% in treated and untreated biocomposites, respectively (Hassaini *et al.* 2017). The gas permeability through samples increased with treated olive husks, with oxygen permeability increasing by 164% and water vapor permeation by 352% (Hassaini *et al.* 2017).

Table 6. Olive Filler-based Biocomposites

Filler	Treatment	Matrix	Preparation Method	Reference
Olive pit powder 5 wt%, 10 wt%, 15 wt%, 20 wt%	Untreated	PLA	Injection molding	Koutsomitopoulou <i>et al.</i> (2014)
Olive husk flour 10 wt%, 20 wt%, 30 wt%	Untreated	PLA	Injection molding	Isadounene <i>et al.</i> (2017)
Olive solid waste 20 wt%	Untreated	PLA and acrylate rubber particles (ACR) wt%, 2.5 wt%, 5 wt%, 10 wt%	Compounding → compression molding	Khemakhem <i>et al.</i> (2018a)
Olive pomace 0 wt% to 20 wt%	Untreated	Plasticized wheat gluten	Compounding → film formation	Boudria <i>et al.</i> (2018)
Surface treated				
Olive husk flour 30 wt%	Acetic or propionic anhydride	PBSA	Compounding → compression molding	Tserki <i>et al.</i> (2005, 2006)
Olive pomace 0 wt% to 20 wt%	Acetone, MA, NaOH	Wheat gluten plasticized by 35% glycerol	Compounding → compression molding	Hammoui <i>et al.</i> (2015)
Olive husk flour 20 wt%	Silane	PHBV	Compounding → compression molding	Hassaini <i>et al.</i> (2017)
Olive solid waste 20 wt%	Hexane	PLA, PCL, and poly(styrene- acrylic-co- glycidyl methacrylate)	Compounding → compression molding	Khemakhem <i>et al.</i> (2018b)
Olive husk flour 20 wt%	NaOH	PLA	Injection molding	Isadounene <i>et al.</i> (2018)

Olive pomace, cake, and waste all refer to the by-product produced from olive oil production. Researchers have investigated the effects of olive pomace in different biopolymer matrixes, including gluten, PLA, and PLA/PCL blend. Hammoui *et al.* (2015) and Boudria *et al.* (2018) investigated the effect of olive pomace in a wheat gluten-based matrix. Hammoui *et al.* (2015) investigated the effects of olive pomace treated with acetone, esterification with maleic anhydride (MA), and mercerization with NaOH. The chemical modifications did not improve the mechanical properties significantly, and the treatments decreased the water absorption of the biocomposites (Hammoui *et al.* 2015). Boudria *et al.* (2018) also incorporated olive pomace with wheat gluten. They found that the moisture absorption was improved (with less moisture

absorption being better for film applications) with increasing addition of olive pomace, compared to neat wheat gluten (Boudria *et al.* 2018), similar to the findings reported by Hammoui *et al.* (2015). They attributed the reduction in water absorption to increased lignin content in the wheat gluten, which would reduce the overall hydrophilicity of the blend (Boudria *et al.* 2018). The difference between the two studies can be explained by the treatment of the olive pomace. Chemical treatments are well known for reducing water absorption because of the changed surface functionalities of the filler. This has been confirmed in other research (Li *et al.* 2007).

It is common for olive-based biocomposites to exhibit decreased tensile strength as compared to neat polymers because the added biofiller/biofiber can introduce stress concentration factors into a polymer matrix. The inefficient stress transfer between the polymer matrix and biofillers/fibers causes the tensile strength to decrease (Fu *et al.* 2008). Some researchers have found that water absorption decreased with the incorporation of treated fibers (Tserki *et al.* 2006), but some reported an opposite water absorption trend (Hammoui *et al.* 2015). The increase in water absorption was explained by an increased amount of lignin present, as lignin is hydrophobic, so the addition of it would decrease water absorption. This work and others with olive pomace have found many creative ways to generate composites; a summary of these works is located in Table 6.

Banana

Banana is an edible fruit produced by different plants from the *Musa* genus. Banana fibers (BF) are a by-product from banana farming and have little value (Merlini *et al.* 2011). Banana plants do not regrow more bananas once harvested, so they generate large amounts of waste biomass (Li *et al.* 2010). The part of the plant that is left over is typically referred to as a “pseudo-stem” (Li *et al.* 2010). The leftover part of the plant is typically allowed to decompose in the field, resulting in environmental pollution. This can be remediated by utilizing the fibers in value-added applications like biocomposites.

Most of the literature on banana biocomposites focuses on treated BF, usually with silane and some with both silane and NaOH. As previously discussed, these treatments help make the fibers less hydrophilic, therefore decreasing water absorption and increasing matrix adhesion between the filler and the polymer in question. Research on BF composites has been focused on the use of PLA as the matrix, with uncompatibilized and compatibilized biocomposites.

Asaithambi *et al.* (2014) investigated the effect of surface treatment with benzoyl peroxide on banana/sisal fibers in PLA (Fig. 6). A fraction of 30 wt% of fibers was blended with 70 wt% of PLA. The authors found that the chemical treatment enhanced the fiber/matrix interaction and increased the tensile strength, flexural strength, modulus, and impact strength, compared to neat PLA. Jandas *et al.* (2011) investigated the effects of surface treatments (washing, NaOH, acetylation, and silane) on the tensile strength of BF. They found that the silane-treated fibers had the greatest tensile strength, modulus, and elongation at break. Acetylation, NaOH treatment, and washing all decreased the tensile modulus and elongation at break. However, the tensile strengths of the samples with different treatments were more variable. The silane-treated samples showed greater tensile strength than the washed samples, followed by the NaOH-treated samples, with the lowest tensile strength belonging to biocomposites with acetylated fibers (Jandas *et al.* 2013c). Jandas *et al.* (2011) also investigated the effect of surface treatment with NaOH and silane on the properties of banana fiber/PLA biocomposites. They found that a

BF loading of 30 wt% yielded optimal mechanical properties. Surface treatment with bis-(3-triethoxy silyl propyl) tetrasulfane (Si69) had the best thermomechanical results (Jandas *et al.* 2011). Surface-treated biocomposites had improvements in both tensile strength and modulus, along with greater glass transition temperature (T_g), crystallization temperature (T_c), and melting temperature (T_m) (Jandas *et al.* 2011). The surface treatment also increased the storage modulus and decreased the degradation rate for samples placed in a *Burkholderia cepacia* and potato dextrose broth. These observations correlate with the enhanced interfacial adhesion and lower microbial activity due to silane's antibacterial activity (Jandas *et al.* 2012). The same group used micromechanical models such as Hirsh's, a modified Bowyer-Bader (Bowyer and Bader 1972), and Brodnyan to model the biocomposites' moduli. Overall, these models showed good agreement with the results obtained, but they predicted the silane treatment more accurately because the treatment improved the interphase between the filler and matrix (Jandas *et al.* 2013a).

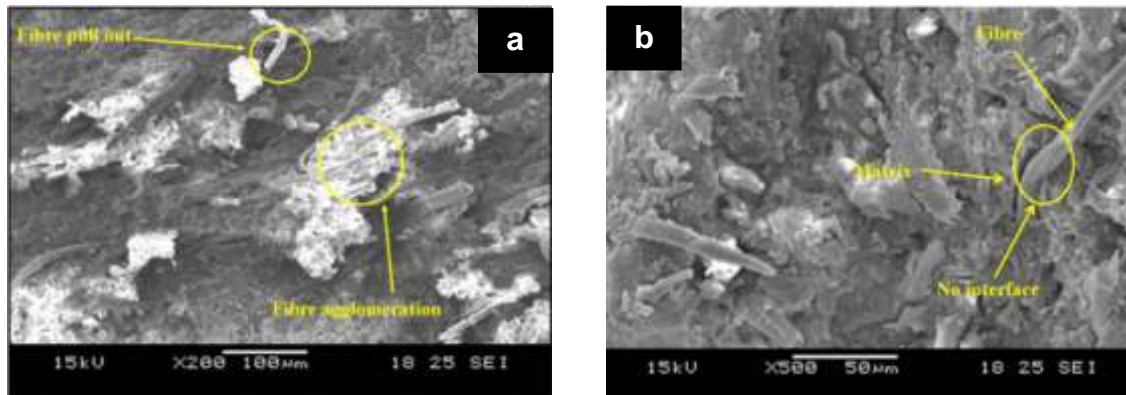


Fig. 6. Banana fibers with PLA: (a) tensile and (b) flexural samples. Reprinted with permission from Springer Nature: *Fibers and Polymers*, (Asaithambi *et al.* 2014), Copyright 2014, License Number: 4561970447709.

Jandas *et al.* (2013b) further investigated the effect of organically modified nanoclay on PLA/surface-treated BF. The incorporation of 3 wt% nanoclay increased T_g from 57.3 °C to 62.6 °C (Jandas *et al.* 2013b), which was attributable to a decrease in free volume in the system, from the cross-linking from the intermolecular attraction between the diols on the nanoclay and the C=O bonds present in the PLA (Tjong and Bao 2005). Thermal stability increased along with the storage modulus. The final degradation temperature of the nanoclay BF composites was 372 °C, which is 30 °C greater than the neat PLA. This increase was attributed to the high thermal stability of the nanoclay and to the interaction between the matrix and BF (Alvarez and Vázquez 2006). The increase in storage modulus was attributed to the large interfacial area and interactions between the PLA and silane-treated BF due to the incorporation of nanoclay. The flammability was decreased to 8.4 mm/min for the nanoclay biocomposites, compared to 10.0 mm/min for the neat PLA. Jandas *et al.* (2013c) took 70/30 PLA/silane-treated BF blend and added 3 wt% nanoclay to make injection-molded samples. The samples were molded cutlery, which had lower density and improved mechanical properties compared to PP or PE. This result demonstrated that the researched biocomposite samples had a potential purpose and an intended application (Jandas *et al.* 2013c).

Majhi *et al.* (2010) combined banana fibers (Fig. 6) with PLA and found that the

tensile strength increased with increasing fiber loadings. However, elongation at break and impact strength decreased with increasing untreated and treated BF (Majhi *et al.* 2010). The BF and PLA showed a noticeable gap between the filler and the matrix, which may explain the decrease in material properties. When 1 wt% of MA was added to the BF/PLA blends, increases in tensile modulus, tensile strength, elongation at break, and impact strength were observed, compared to their uncompatibilized counterparts. The authors also found that increasing the MA content to 3 wt% or 5 wt% caused the tensile modulus to reach a plateau, which could be considered a critical interfacial concentration, beyond which no change in properties would be seen with additional amounts of compatibilizer (Liu *et al.* 2003).

Sajna *et al.* (2016) created biocomposites with PLA and PLA-grafted (PLA-g) glycidyl methacrylate (GMA) as a compatibilizer with treated BF. Silane-treated BF at 30 wt% loading was incorporated with 70 wt% PLA. Then, various weight percentages (5 wt%, 10 wt%, 15 wt%, and 20 wt%) of PLA-g GMA were added (Sajna *et al.* 2016). The compatibilized biocomposites showed improvements in their mechanical properties compared to neat PLA and their uncompatibilized counterparts. The degradation temperature improved with GMA, along with the moisture absorption resistance. The researchers then incorporated nanoclay into the PLA/silane-treated BF matrix without the GMA. The nanoclay increased the thermal degradation temperature compared to the neat PLA and the biocomposites without the nanoclay (Sajna *et al.* 2017a). The thermal stability was improved, and the combustion rate decreased, which was attributed to the fiber and nanoclay producing char, acting as a thermal barrier (Sajna *et al.* 2017a). The researchers then explored the effect of nanoclay on compatibilized biocomposites with GMA (10 wt%, 15 wt%, 20 wt%, and 25 wt%) and nanoclay. The thermal stability of the biocomposites was further enhanced with the addition of GMA. The compatibilized composite showed the slowest burning rate, at 13.1 mm/min, compared to 15.8 mm/min for neat PLA (Sajna *et al.* 2017b). Overall, it was determined that silane-treated BF with the incorporation of nanoclay and compatibilized PLA created the best biocomposites in terms of mechanical and thermal properties.

Banana biocomposites created by Asaithambi *et al.* (2014) showed improved mechanical properties compared to those of the neat polymer. In fact, there were 175%, 400%, 158%, 151%, and 106% increases in tensile strength, tensile modulus, flexural strength, flexural modulus, and impact strength, respectively, for the treated BF composites (Asaithambi *et al.* 2014). Jandas *et al.* (2015) investigated various different BF compositions and noted that surface treated fibers showed improvement compared to the untreated filler. They also noted that the thermal stability improved with the addition of nanoclay (Alvarez *et al.* 2004; Jandas *et al.* 2013b). The addition of compatibilizers, MA and GMA, respectively, also improved the mechanical properties of banana biocomposites (Majhi *et al.* 2010; Sajna *et al.* 2016).

Banana skin has been made into flour and combined in varying weights (approximately 8 wt% to 32 wt%) with PVOH, HTMA, glycerol, and polysorbate 80 to produce sustainable films. Some of the biofilms' properties were hindered by the addition of banana skin flour, showing decreased tensile strength, increased water absorption, and increased water vapour transmission rate (Zhong *et al.* 2011). Uniquely, Zhong *et al.* (2011) examined the natural weathering characteristics of the films, with natural weathering analysis of samples subjected to environmental factors such as sunlight, rain, dew, and wind. They found that, although the tensile strength and elongation at break decreased after 1 month of weathering, the tensile moduli of blends with banana skin

flour increased. This result was attributed to the breaking down of long molecular chains in the PVOH materials (Zhong *et al.* 2011). A summary of the processes to fabricate these composites and others from banana can be found in Table 7.

Table 7. Banana Fiber-based Biocomposites

Filler	Treatment	Matrix	Preparation Method	Reference
BF 10 wt%, 20 wt%, 30 wt%, 40 wt%	NaOH	PLA, MA, and GTA	Compounding → compression molding	Majhi <i>et al.</i> (2010)
BF 10 wt%, 20 wt%, 30 wt%, 40 wt%	NaOH and silanes	PLA	Compounding → compression molding	Jandas <i>et al.</i> (2011, 2012, 2013a, 2013c)
BF 30 wt%	Washed in detergent, NaOH, silane	PLA and organically modified nanoclay	Injection molding	Jandas <i>et al.</i> (2013b)
BF 30 wt%	Benzoyl peroxide	PLA	Injection molding	Asaithambi <i>et al.</i> (2014)
BF 5 wt%, 10 wt%, 15 wt%, 20 wt%	Silane	PLA and PLA-g GMA	Injection molding	Sajna <i>et al.</i> (2016)
BF 30 wt%	Silane	PLA/ 3 wt% nanoclay	Injection molding	Sajna <i>et al.</i> (2017a)
BF 20 wt%	Silane	PLA, PLA-g GMA, and nanoclay 3 wt%	Injection molding	Sajna <i>et al.</i> (2017b)
Banana skin 8 wt% to 32 wt%	Untreated	PVOH, glycerol, HMTA, polysorbate 80	Film	Zhong <i>et al.</i> (2011)

Coconut

Coconut (*Cocos nucifera*) is a member of the palm tree family and has edible fruit. Coconut fibers (CF), or coir, are located in the middle of the husk and the outer shell in a coconut. The price of the fibers is low, as they are a by-product of other coconut products such as coconut milk and flesh. Out of all coconuts produced, it is estimated that approximately 10% of the husks are used as fibers (Faruk *et al.* 2012). The majority of research on coconut biocomposites focuses on PLA, PBS, EVOH, polyester amide, and gluten polymers.

Nam *et al.* (2011) incorporated coir fibers into PBS at different weight percentages, between 10 wt% and 30 wt%, of filler. The fibers were treated with NaOH before being compression molded with PBS. A ratio of 75 wt% to 25 wt% of PBS to CF showed the best mechanical properties, with increases of 54.5% for tensile strength, 141.9% for tensile modulus, 45.7% for flexural strength, and 97.4% for flexural modulus, when compared to the neat PBS (Nam *et al.* 2011). The authors concluded that up to 25 wt% of fibers could be incorporated in a PBS matrix before the mechanical properties started to decrease.

Rosa *et al.* (2009) incorporated 15 wt% coir into starch/EVOH blends at 50 wt% starch, 30 wt% EVOH, 10 wt% water, and 10 wt% glycerol. They performed water, alkali, and bleaching treatments on the CF. The treated fibers had greater tensile strength than those untreated and showed greater tensile strength and tensile modulus than

starch/EVOH, along with greater thermal stability (Rosa *et al.* 2009). The authors concluded that the addition of CF into the biocomposite yielded enhanced material properties compared to the neat polymer.

Hemsri *et al.* (2012) created wheat gluten composites with 15 wt% CF treated by alkali or alkali and silane and 85 wt% wheat gluten. The silane-treated biocomposites had better properties than the alkali-treated CF biocomposites. The addition of treated fibers increased the stress at the first failure point on the stress-strain curve, and fiber treatment improved the adhesion with the matrix material, reducing the fiber pull-out observed in the uncompatibilized samples (Hemsri *et al.* 2012).

Rout *et al.* (2001) made CF and polyester amide biocomposites. Four surface treatments were evaluated including alkali, cyanoethylation, bleaching, and vinyl grafting. The optimal fiber content was 50 wt%, in terms of mechanical properties. All of the surface treatments improved the mechanical properties compared to neat polymer, with the cyanoethylation treatment yielding the greatest tensile strength (Rout *et al.* 2001). Biocomposites with 7% methyl methacrylate grafted coir had superior mechanical properties in comparison to the other composites (Rout *et al.* 2001).

Suardana *et al.* (2011) fabricated biocomposites with 40 wt% CF and 60 wt% PLA. Coconut fibers were rinsed, treated with NaOH followed by acrylic acid, and then rinsed. The biocomposites treated with acrylic acid had the least water absorption compared to untreated or only NaOH-treated fibers (Suardana *et al.* 2011). Soaking in acrylic acid for 0.5 h yielded the greatest tensile and flexural strengths of the biocomposites after 3 h, 6 h, and 192 h of water immersion (Suardana *et al.* 2011). Salmah *et al.* (2013) also used acrylic acid treatment to modify the surface of coconut shell powder. They found that the treated biocomposites had greater tensile strength and modulus of elasticity, but the elongation at break was reduced (Salmah *et al.* 2013). The surface treatment was found to enhance the matrix-filler interaction and increase the thermal stability.

Jang *et al.* (2012) created biocomposites with oxygen plasma-treated CF and PLA. Fractions of 5 wt% and 10 wt% untreated and treated CF were incorporated into a PLA matrix. The tensile strength and Young's modulus increased, and shrinkage of the biocomposites decreased with increasing fractions after treatment of the fibers (Jang *et al.* 2012). Dong *et al.* (2014) treated CF with NaOH and created CF/PLA composites. They found that 20 wt% CF yielded the optimal balance of mechanical properties. The alkali treatment improved all mechanical properties compared to the untreated fiber, which the authors attributed to the improved fiber-matrix adhesion and fiber wettability (Dong *et al.* 2014). They also found that the thermal stability of the biocomposites decreased when treated, due to the lower thermal stability of the fibers incorporated. The addition of the fibers increased the degradation rate, which the authors attributed to the residual NaOH causing breakup of the PLA chains, in turn leading to faster degradation (Dong *et al.* 2014).

Some researchers have found that the incorporation of CF into various biopolymer matrices yielded an improvement in certain mechanical properties (Nam *et al.* 2011; Suardana *et al.* 2011). Thermal stability was also seen to improve with the addition of treated fibers (Rosa *et al.* 2009; Salmah *et al.* 2013). The addition of compatibilizer further improved the mechanical properties (Rout *et al.* 2001), and a decrease in shrinkage was observed with increasing weight percentages of CF (Jang *et al.* 2012). A greater degradation rate in a composting medium was seen when CF was introduced into a PLA matrix, indicating improved composting. This was attributed to the

increased water diffusion through the sample due to the hydrophilicity of the fillers (Dong *et al.* 2014). A summary of all biocomposites generated from coconut is displayed in Table 8.

Table 8. Coconut Filler- and Fiber-based Biocomposites

Filler	Treatment	Matrix	Preparation Method	Reference
Coconut shell powder, 15 wt%, 30 wt%, 45 wt%, 60 wt%	Silane	PLA	Compounding → compression molding	Chun <i>et al.</i> (2012)
Surface treated				
Coir, 30 wt%, 40 wt%, 50 wt%, 60 wt%	NaOH, bleaching	Biodegradable polyester amide	Sandwiched mats, compression molded	Rout <i>et al.</i> (2001)
Coir, 15 wt%	NaOH, bleaching	EVOH, starch, water, glycerol	Injection molding	Rosa <i>et al.</i> (2009)
Coir, 40 wt%	NaOH and acrylic acid	PLA	Compression molding	Suardana <i>et al.</i> (2011)
Coir, 10 wt%, 15 wt%, 20 wt%, 25 wt%, 30 wt%	NaOH	PBS	Compression molding	Nam <i>et al.</i> (2011)
CF 15 wt%	NaOH, silane	Wheat gluten	Compression molding	Hemsri <i>et al.</i> (2012)
Coir 5 wt%, 10 wt%,	Treated	Oxygen plasma/PLA	Compression molded	Jang <i>et al.</i> (2012)
Coconut shell powder, 0 wt%, 15 wt%, 30 wt%, 45 wt%, 60 wt%	Untreated and acrylic acid	PLA	Compression molding	Salmah <i>et al.</i> (2013)
Coir, 5 wt%, 10 wt%, 20 wt%, 30 wt%	NaOH	PLA	Compression molding	Dong <i>et al.</i> (2014)

Pineapple

Pineapple (*Ananas comosus*) is an abundant tropical plant that generates a lot of lignocellulosic fibers (Fig. 7). Currently, pineapple leaf fibers (PALFs) are considered a by-product of pineapple farming, so they can be obtained at a low cost (Faruk *et al.* 2012). Pineapple leaf fiber biocomposites show the most variability in terms of surface treatments and matrices out of all the biofillers/fibers explored. Untreated, NaOH-treated, and silane-treated PALFs have been added to different matrices, including PLA, PBS, polyester amide (PEA), soy flour, and tapioca.

Kim *et al.* (2012) created pineapple flour/PLA biocomposites. The flour was blended with MA-grafted PLA (MA-g PLA), with 3 wt% MA-g PLA incorporated with different weight percentages of filler. It was found that 30 wt% of flour yielded the best overall mechanical properties (Kim *et al.* 2012). Furthermore, T_g increased with the addition of flour, while T_m was divided into two peaks, likely due to the quick decrease in molecular weight and rearrangement of polymer chains (Kim *et al.* 2012). The heat deflection temperature also increased with the flour and MA, from 56.8 °C to 70.6 °C, indicating improved stress transfer (Kim *et al.* 2012).

Liu *et al.* (2005) created soy flour, PEA, and chopped PALF composites. The flour was mixed at a 70:30 weight ratio to PEA and then extruded with 15 wt% or 30

wt% PALF. The 70:30 blend with 30 wt% PALF was then combined with a compatibilizer at 5 wt% (PEA grafted with GMA (Liu *et al.* 2005). The addition of 30 wt% PALF significantly increased the mechanical properties. The improvements were even more pronounced with the addition of compatibilizer. The water absorption of the compatibilized samples was lower than that of the uncompatibilized counterparts, which was attributed to the improvement of interfacial adhesion and stress transfer between the matrix and fiber (Liu *et al.* 2005).

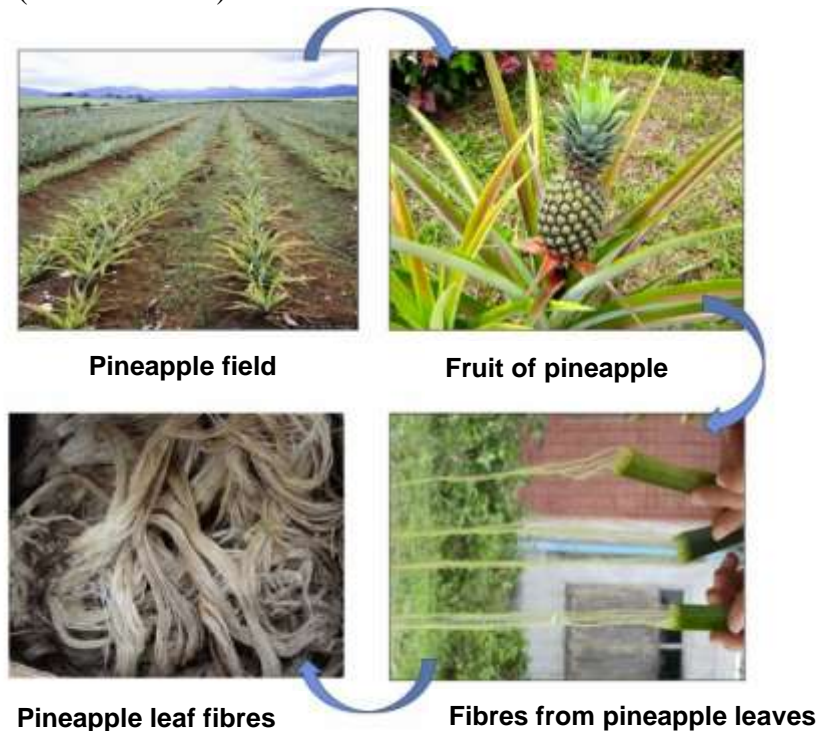


Fig. 7. Pineapple field, fruit, and leaf fibers (Asim *et al.* 2015)

Siakeng *et al.* (2019) fabricated coconut/pineapple fiber and PLA biocomposites (Fig. 8). They incorporated a 1:1, 3:7, or 7:3 ratio of coconut to pineapple at 30 wt% or 30 wt% of either into a PLA matrix. The authors found that the hybrid combinations of the fibers performed better than the individual fiber types alone (Siakeng *et al.* 2019). The authors attributed this result to the combination of fibers overcoming the inherent disadvantages of each of the fiber types individually. The addition of CF decreased tensile and flexural strength, while the opposite effect was seen with the addition of PALF (Siakeng *et al.* 2019). A ratio of 15 wt% CF with 15 wt% PALF and 70 wt% PLA had the best tensile and flexural properties, along with the storage modulus and loss modulus.

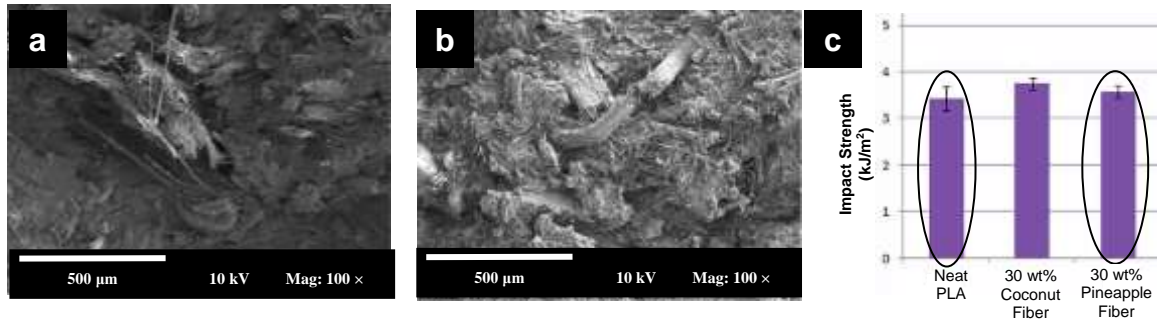


Fig. 8. Pineapple fiber composites: (a) tensile fracture surface, (b) flexural fractural surface, (c) impact properties where the error bar is the standard deviation. Reprinted with permission from John Wiley & Sons: *Polymer Composites*, (Siakeng *et al.* 2019), Copyright 2018, License Number: 4557640115394.

Jaafar *et al.* (2018) fabricated PALF (at 10 wt%, 20 wt%, 30 wt%, and 40 wt%) and PLA biocomposites. The optimal percentage of filler was 30 wt%, with improvements in tensile strength, tensile modulus, flexural strength, and impact strength of 42%, 165%, 69%, and 10%, respectively (Jaafar *et al.* 2018). Huda *et al.* (2008) examined the effects of untreated and treated PALF fibers in a PLA matrix. They performed alkali, silane, and combined alkali/silane surface treatments on the PALF. Biocomposites of 30 wt%, 40 wt%, and 50 wt% PALF were created. The biocomposites created from the treated fibers had significantly improved mechanical and thermal properties (Huda *et al.* 2008). These increases were attributed to the strong interaction between the added PALF and the PLA. Dynamic mechanical analysis revealed a greater storage modulus, indicating greater interfacial bond strength and fiber matrix adhesion. This result was attributed to the chemical bonds between the PALF and the matrix due to the reactive groups present on the fiber surface (Huda *et al.* 2008).

Shih *et al.* (2014) fabricated treated PALF and recycled chopstick (wood) fibers in a PLA or PBS matrix. The fibers were treated with an alkaline solution and a silane treatment. The fibers at 30 wt% (7:3 ratio of PALF to chopstick: 21 wt% PALF and 9 wt% chopstick) yielded the best overall mechanical properties. The tensile strengths of the PBS and PLA increased by 121.7% and 66.1%, respectively (Shih *et al.* 2014). The flexural strengths increased for both PBS and PLA by approximately 66%, and the heat deflection temperatures of the PBS and PLA increased by approximately 33.6% and 75%, respectively (Shih *et al.* 2014).

Ramli *et al.* (2016) created PALF and PLA biocomposites with both treated and untreated PALF with long and short fibers. With the long fibers, the flexural strength and modulus increased from 56.4 to 114 MPa and 4.3 to 5.7 GPa when treated, respectively, with 30 wt% loading. The authors attributed this increase to the alkaline treatment increasing the surface roughness of the fibers, causing a rough surface at the matrix-fiber interface (Ramli *et al.* 2016).

Table 9. Pineapple Fiber-based Biocomposites

Filler	Treatment	Matrix	Preparation Method	Reference
PALF 40 wt%	NaOH and silane	PLA	Compression molding	Huda <i>et al.</i> (2008)
Pineapple skin flour 30 wt%	Untreated	PLA and MA-g PLA	Injection molding	Kim <i>et al.</i> (2012)
PALF/chopstick fiber 10 wt% to 30 wt%	NaOH	PBS and dicumyl peroxide	Injection molding	Shih <i>et al.</i> (2014)
PALF 15 wt%, 30 wt%	Untreated	Soy flour, PEA, and PEA-grafted GMA	Injection molding	Liu <i>et al.</i> (2005)
PALF 30 wt %	Untreated and NaOH	PLA	Compounding → compression molding	Ramli <i>et al.</i> (2016)
PALF/CF (1:1, 7:3, 3:7) 30 wt%, CF 30 wt%, PALF 30 wt%	Untreated	PLA	Compounding → compression molding	Siakeng <i>et al.</i> (2019)
PALF 10 wt%, 20 wt%, 30 wt%, 40 wt%	Untreated	Tapioca biopolymer	Compounding → compression molding	Jaafar <i>et al.</i> (2018)

The addition of PALF to a polymer matrix improved the mechanical properties, compared to a neat biopolymer (Jaafar *et al.* 2018). The mechanical properties were further improved with the addition of compatibilizer into the PALF/biopolymer system (Liu *et al.* 2005; Kim *et al.* 2012) or with surface treatment of the PALF (Huda *et al.* 2008; Ramli *et al.* 2016). The heat deflection temperatures of the biocomposites increased with compatibilizer or surface treatment (Kim *et al.* 2012; Shih *et al.* 2014). Siakeng *et al.* (2019) found that a combination of both CF and PALF yielded better mechanical properties than either of the fiber types individually (Siakeng *et al.* 2019). This could be an interesting area of further research, combining fibers based on their strengths and weaknesses to develop composites with desired properties. A summary of this work and all works with pineapple fiber is shown in Table 9.

ENVIRONMENTAL CONSIDERATIONS

There are different considerations regarding the environmental impacts of agricultural and food wastes. Some environmental concerns are related directly to waste generation, such as composting, incineration, or landfilling. Other impacts from the agri-waste correspond to the secondary processing for chemicals or fuels. It is essential to consider the effects on soil, air, and waterways from waste products and greenhouse gas emissions.

Current Uses of Fruit Waste

Work by Khan *et al.* (2015) highlights various uses for fruit wastes (Fig. 9). Although their work focuses on South Africa, similar processes are applicable around the world. A sustainable bio-economy requires the use of materials in industrial applications such as bio-materials, bio-chemicals, and bio-energy, as well as sustainable environmental management processes for liquid and solid wastes (Khan *et al.* 2015).

Environmental Impact

Fruits are a major factor in the agri-food processing industry. In the United States of America, the food processing industry requires one fifth of all energy consumed (Diakosavvas 2017). Furthermore, a substantial amount of energy is put forth to transport the materials from farms to processing facilities and then to their final destinations, as well as for refrigeration of the fruits (Diakosavvas 2017). Therefore, it is important to maximize the use of the residue or pomace materials. A reduction in energy consumption should be followed by an overall decrease in greenhouse gas emissions (Diakosavvas 2017).

Traditional methods of discarding fruit waste include placing the material in landfills, use as a feed source for livestock, and compositing (Negro *et al.* 2017). Some pomace materials are further refined for chemical extracts, such as pectin from apples (Shalini and Gupta 2010). Other methods of discarding waste are resource regenerative methods, including thermomechanical and biological energy production. The thermomechanical processes use pyrolysis or incineration of the materials to generate heat and electricity; biological processes such as anaerobic digestion and co-digestion produce bio-products and fuel (Negro *et al.* 2017). However, some of these processes can yield harmful by-products such as carbon dioxide and methane, which may contribute to greenhouse gas emissions (Negro *et al.* 2017).

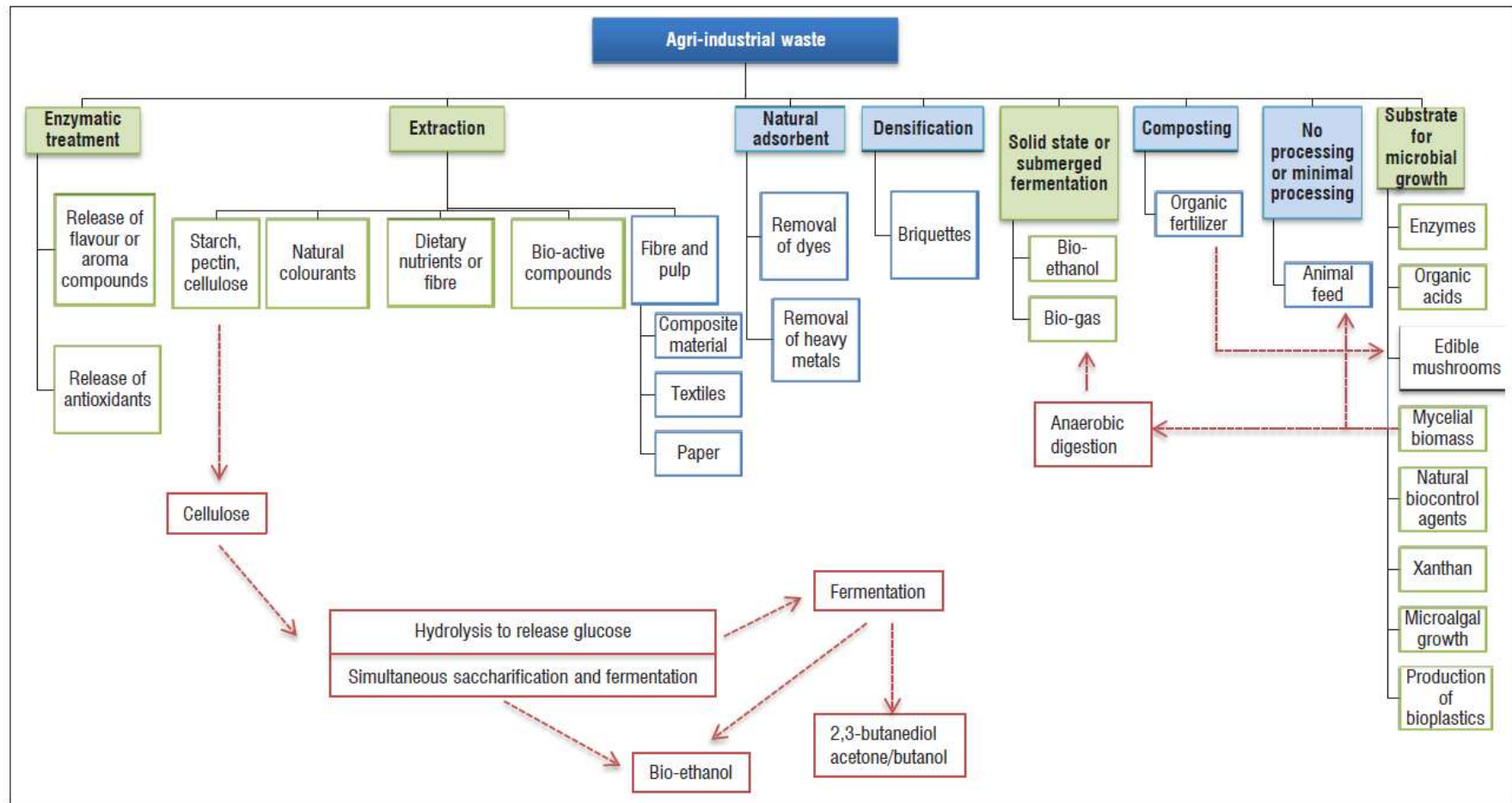


Fig. 9. Description of agri-waste (with major focus on fruit) uses and management systems in South Africa (Khan *et al.* 2015)

Feasibility of Manufacturing

To ensure valorization of fruit waste, optimal methods to store and process the materials are required so that they are accessible all year round. Some fruit wastes are generated at the time of harvest, while others are generated throughout the year. Olive and grape pomaces, for example, are available year-round as they remain in storage prior to processing, thus providing steady supply of this material for value-added applications (Gowman *et al.* 2019). Other fruit such as bananas are produced year-round and therefore are considered as a continuous source of fruit waste (Asim *et al.* 2015).

Once fruit wastes, such as apple and grape pomaces are generated, they are stored temporarily in a cold cellar to reduce the rate of spoilage and then dried as soon as possible (Gowman *et al.* 2019). The materials can be air-dried if aerated frequently, which is a cost-effective drying method. Wastes are typically dried to a moisture content of less than 1 wt% and sealed in cool storage to minimize spoilage. This approach ensures that the materials can be transported and utilized in industry with ease.

Life Cycle Assessment for Fruit Wastes

A literature review by Gassara *et al.* (2011) compared disposal methods for AP via a life cycle assessment. Comparing AP from Quebec, Canada, destined for incineration, landfill, fermentation, composting, or animal feed (Table 10), the authors determined that fermentation contributed the least to emissions, in terms of CO₂ equivalence (Gassara *et al.* 2011). The addition of AP or natural fibers prolongs the life of the sample and does not result in immediate placement of the material into one of the categories mentioned below. In fact, the use of fruit waste in composites sequesters the carbon and prevents the release of carbon dioxide until the biocomposites and films are at the end of their functional lives. The end of life of the materials would most likely result in landfill, but the materials will degrade naturally and more easily with greater natural filler content. However, there are other methods of disposal.

Table 10. Comparison of Carbon Emissions from Various Disposal Streams of AP in Canada

Fruit	Incineration (tonnes of CO ₂ equivalent)	Landfill (tonnes of CO ₂ equivalent)	Fermentation (tonnes of CO ₂ equivalent)	Composting (tonnes of CO ₂ equivalent)	Animal Feed (tonnes of CO ₂ equivalent)	Location
AP Gassara <i>et al.</i> (2011)	1017.95	1670.13	822.64	1155.55	873.96	Quebec, Canada

According to the literature, if the biobased polymers are burned at the end of life, then the sample is considered carbon neutral. Carbon neutrality occurs because the carbon dioxide generated from burning the materials is then used by the biomass during photosynthesis (Iwata 2015). Therefore, valorization of the waste product creates energy-conscious products which are sustainable for years to come.

Life Cycle Assessment for Fruit Waste Composites

It is important to reiterate that the biodegradability of non-biodegradable polymer is not affected by the addition of natural fillers or fibers. Although there is improved

sustainability from the production of composites, those with non-biodegradable matrix material most likely would end up in incineration or landfill at their end of life, as recycling of these materials is not feasible. The more ideal option is to combine natural fillers from fruit wastes with biodegradable polymers (Mohanty *et al.* 2018). This approach alleviates the risk of contaminating the recycling streams, and allows for industrial composting.

CONCLUSIONS

1. Fruit wastes have the potential to be successfully incorporated into many different biodegradable polymer matrices for a wide variety of applications. The use of fruit waste helps valorize the by-product, generating revenue for farmers and lower material costs for composite manufacturers.
2. These biocomposites have the advantage of being biodegradable and lower cost than their neat biopolymer counterparts, due to the incorporation of the low-cost biofiller/fiber.
3. A major hurdle to overcome is the creation of biocomposites having a good balance of mechanical and thermal properties, while maintaining the low-cost benefits associated with the fruit waste. Surface treatments of biofillers/fibers enhance these properties, but a greater cost is associated with these treatments. Compatibilizers have also been effective in improving mechanical and thermal properties, but they also have a high associated cost.
4. Further work on utilizing fruit waste biocomposites in real-life applications would help advance the field as well as the impact of these materials on the environment. Valorization of fruit waste biomass in biocomposites will continue to gain favor in industry and research, as the materials provide a renewable and sustainable solution to global plastic concerns.

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