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Review

Upcycling Food Waste into Biomaterials Applicable to Medical Products

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Abstract: Globally, an estimated 1.3 billion tons of food are wasted each year, according to a report from the Food and Agriculture Organization of the United Nations. A variety of waste streams constantly generate large amounts of food waste that end up in landfills. As food waste is left to naturally decay in landfills, it emits greenhouse gases that pollute the environment and induce climate change. However, most types of food waste contain valuable components that can be extracted to manufacture industrial products. Therefore, instead of abandoning food waste to decay and harm the environment, there is an alternative to upcycle it as a new raw materials supply source. This review provides a comprehensive update on how environmental sustainability can be improved using diverse types of food waste as sources to generate biomaterials for fabricating medical products, including lignin, cellulose, chitosan, pectin, collagen, hydroxyapatite, and biodegradable polymers. The review also highlights biochemical technologies applied for extracting useful components from food waste and details the current advances for developing medical products, including wound dressings and nanoparticles for tissue engineering and drug delivery.

Keywords: food waste; bioconversion; upcycle; environmental biotechnologies; medical materials

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1. Introduction

Food waste (FW) is food that is not used for a specific purpose and is, hence, disposed of. FW generated from various waste streams has become an increasingly severe issue. There is an estimated 1.3 billion tons per year of FW across the globe, with the main contributors being industrial, consumer, and retail waste streams [1]. These FWs end up in landfills with no practical uses. According to the U.S. Environmental Protection Agency (EPA), the U.S. discards nearly 60 million tons of FW every year, making up 22% of municipal solid waste in landfills. FW deterioration in landfills then emits the greenhouse gases methane and carbon dioxide, which are known to cause environmental pollution, global climate changes, and extreme weather (Figure 1A). They also disrupt the surrounding soil ecosystem via resource depletion [2,3]. In fact, 58% of fugitive methane emissions at landfills come from wasted food, according to an updated report from the U.S. EPA [4].

One strategy to reduce the impact is for retailers in the food supply chain to improve efficiency and reduce FW while allowing for increased savings, employment, and competition [3]. A second strategy is to upcycle FW. This has been a trending topic across many fields, trying to repurpose the waste into useable products. For instance, FW can be used in energy production to create biodiesel, bioethanol, biobutanol, biogas, etc. [5,6]. Similarly, ways are being explored to produce biomaterials by extracting a variety of biopolymers and fabricating bioplastics from FW [7]. This review provides a comprehensive update on how diverse types of FW can be transformed into biomaterials for fabricating medical products, including lignin, cellulose, chitosan, pectin, collagen, hydroxyapatite, and biodegradable polymers. This review also highlights biochemical technologies applied for

extracting useful components from FW and details the utilities of FW-derived materials for developing medical products, including wound dressings and nanoparticles for tissue engineering and drug delivery.

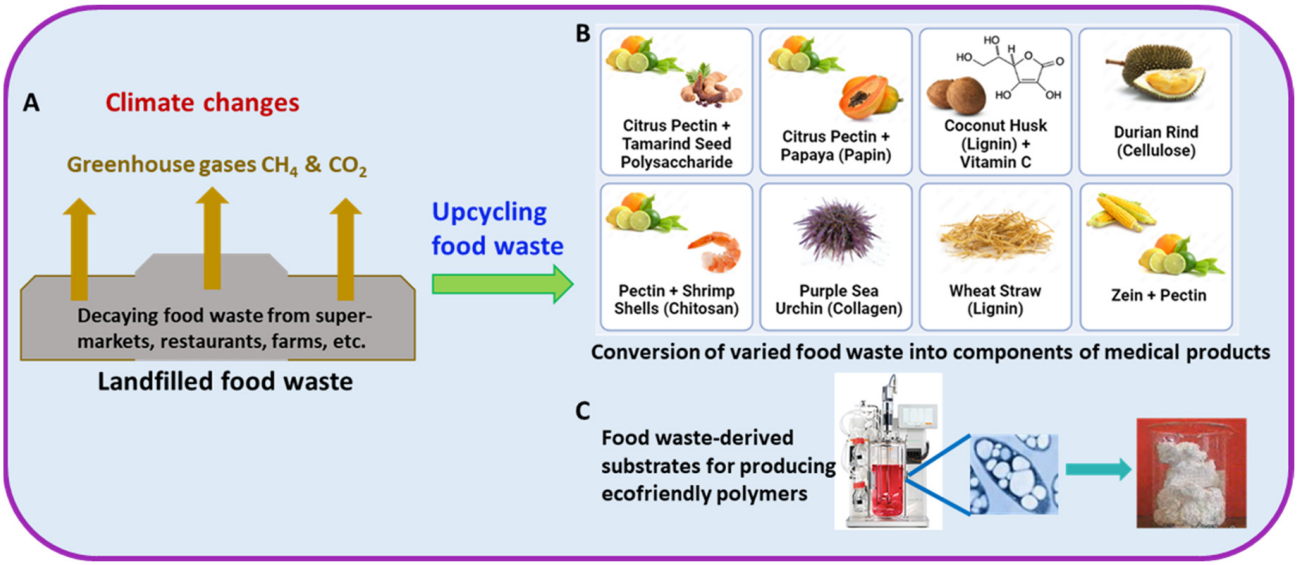

Figure 1. (**A**) The decay of landfilled food waste causes the emissions of greenhouse gases, including warming power gases methane and carbon dioxide in the atmosphere, which is responsible for climate change. (**B**) Valuable components can be extracted from a variety of food waste for developing medical products. (**C**) Food waste bioconversion into ecofriendly polymers that have great utilities in making medical products.

2. Food Waste as a Source for Making Valuable Biomaterials

Various materials can be extracted from FW as vital biomaterials. Materials such as lipids, lignin, hemicellulose, cellulose, pectin, etc., can be generated from plant FW. Through pretreatments and hydrolysis, these FWs are converted to sugars that can be used to produce bioplastics, bioethanol, and other high-value materials. Animal-derived FW, on the other hand, is often an excellent source of collagen, chitosan, hydroxyapatite, enzymes, nutrients, and other molecules. Some FW-derived materials are more abundant and have good qualities, such as strength and a hydrophilic nature, that make them ideal candidates for developing medical products. The materials that can be generated from FWs, including lignin, cellulose, chitosan, collagen, pectin, hydroxyapatite, and biodegradable plastics, are discussed in detail herein.

2.1. Lignin Extraction from Food Waste

Lignin is a polyphenolic polymer and one of the main components in the plant cell wall known for providing strength and rigidity to materials. It is formed by the repetition of three main units in a varied sequence [8]. The main building blocks that form the primary structure are p-coumaryl alcohol, coniferyl alcohol, and sinapyl alcohol, with varying end groups (Figure 2).

Currently, lignin's industrial value is growing. It can be used for automotive brake pads, wood panel products, polyurethane foams, and epoxy resins for printed circuit boards [9–12]. Recently, it has also become a material of interest for producing medical products, as lignin is highly non-toxic, has good cytocompatibility, and is biodegradable [13,14]. Additionally, its phenylpropane structures have a strong antioxidant property (Figure 2) [15].

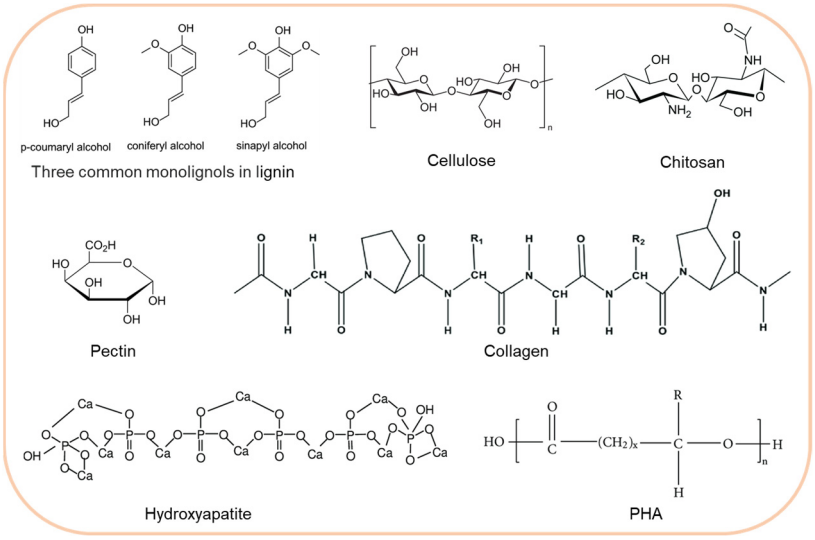

Figure 2. The structural formulae of the biomaterials that can be extracted from food waste or produced from microorganisms.

There are a variety of studies on how to extract lignin. Athinarayan et al. extracted lignin and silica from rice husk using a hydrochloric acid pretreatment, followed by a sodium hydroxide solution, and then an alkaline solution to solubilize the products [13]. In another study, lignin was extracted from wheat straw using an adopted alkali extraction method in which the grounded wheat straw was pretreatment with a toluene and ethanol mixture and then a sodium hydroxide solution, after which precipitated lignin was separated using repeated precipitation and collected [14]. Lignin can also be extracted from wheat straw using a hydrothermal pretreatment and enzymatic hydrolysis, followed by drying to be used in an aerogel formulation [16]. Furthermore, Xu et al. extracted lignin from coconut husks using a mixture of ethanol, deionized water, and sulfuric acid [15]. The resultant mixture was heated and stirred at 70 °C for 4 days, leading to the production of highly pure lignin [15]. Jackfruit pulp and peels, too, have been recycled for lignin extraction through physical pretreatment of ball milling, sulfuric acid treatment, and microwave irradiation [17]. The researcher then used the resultant lignin to make a wound dressing [17]. Moreover, lignin could be extracted from corn cobs using a hydrothermal treatment followed by an alkaline treatment [18]. The alkali lignin was able to self-assemble to form nanospheres that carry bioactive molecule resveratrol for efficient drug delivery [18]. As of today, the most common and effective methods of extracting lignin from plant-based FW include a physical, chemical, or enzymatic pretreatment to breakdown the substrate, followed by removing unwanted portions, such as cellulose and hemicellulose, via separation through filtration or precipitation.

2.2. Cellulose Extraction from Food Waste

Cellulose can be used in making paper, building wood, clothing, and food industries [19–22]. It is a polymer chain consisting of beta acetal-linked glucose units with a formula of $C_6H_{10}O_5$ [23]. Cellulose is one of the components in medicine for wound healing and pain relief. To assess the feasibility of upcycling, Piccinno et al. used carrots as a substitute to model industrial-sized vegetable waste streams. They fitted the life cycle assessment, where carrot waste was depolymerized using an enzyme blend to remove the cellulose [24]. Alternate options have been explored with the durian fruit, which is a good candidate because less than half of the entire fruit is edible, leaving parts such as the rind and seeds to accumulate as FW [25]. To deal with the accumulated waste, Cui et al. adopted a method for extracting cellulose from durian rind to construct an organohydrogel. Durian rind was collected, freeze dried, and ground up using a ball milling technique to create a fine powder. The resultant powder was then treated with pectinase, a 5% NaOH, followed

by a 2% surfactant, and lastly 1% bleach. The final suspension was washed and freeze-dried to produce durian rind cellulose powder (Figure 1B) [25]. In another study, Sommano et al. extracted cellulose from coffee pulp using a pretreatment of dichloromethane and ethanol to remove fat-soluble components [26]. Then, fiber was removed using ammonium oxalate, followed by a purification step using hydrogen peroxide and sodium borohydride to remove lignin, resulting in cellulose substrate. The cellulose, together with pectin and alginate, was fabricated into hydrogels that are non-toxic to a human keratinocyte cells HaCaT [26], suggesting the medical utility of the FW-derived materials.

2.3. Chitosan Extraction from Food Waste

Chitin is a cellulose-like biopolymer that is present in the exoskeletons of crustaceans and insects and the cell walls of fungi and yeast [27]. It is a polymer composed mainly of (1-4)-linked 2-acetamido-2-deoxy-β-d-glucose monomers. Chitosan is the deacetylated derivative of chitin and is composed of (1-4)-linked 2-amino-2-deoxy-β-D-glucose monomers [27]. Chitosan has extensive utilities in medicine, pharmaceuticals, food, cosmetics, agriculture, and paper, as well as the energy industry [28–31]. In particular, chitin and chitosan can be applied in drug delivery, tissue engineering, wound dressings, etc. [32]. Boric et al. evaluated a dielectric barrier discharge method as a pretreatment for shrimp shells to allow for better chitin isolation (Figure 1B) [33]. This method offers thermally non-equilibrium plasmas with a much higher electron temperature [34]. It demonstrated an efficient and fast degradation of proteins from shrimp shell waste, resulting in the extraction of α-chitin [33]. Chakravarty et al. utilized lobster shell waste to extract chitin through either a chemical treatment or biological treatment. The chemical treatment compared two different methods: (1) deproteinization with sodium hydroxide followed by demineralization with hydrochloric acid, and (2) demineralization with sodium hydroxide followed by deproteinization with hydrochloric acid [35]. A biological treatment was carried out by inoculating a coculture of either *Bacillus megaterium* or *Serratia marcescens* with *Lactobacillus plantarum*. They obtained a final chitin yield of 82.56% from the lobster shell biomass [35]. Mohammed et al. used sodium hydroxide and hydrochloric acid for deproteination and demineralization, followed by decolorization using acetone for collecting chitin from prawn shells [36]. The authors also performed deacetylation of the chitin using sodium hydroxide and found that about 25% of the original waste can be converted to chitosan [36]. Arafat et al. used shrimp shell waste to obtain chitosan through demineralization of the shells with HCl, then deproteinization with NaOH, and lastly deacetylation with sodium hydroxide [37]. They obtained 19% of chitosan indicating that shrimp shell is a good source of chitosan. Ghorbani et al. utilized chitosan derived from shrimp shells or pectin derived from citrus peels together with cellulose nanocrystals to fabricate hydrogels. The hydrogels showed enhanced mechanical strengths and were injectable. Furthermore, the authors noted that an increased cellulose content made the hydrogel more resistant to degradation and resulted in increased chondrocyte proliferation, showing that the hydrogel can support cell growth [38]. These studies suggested that the use of bacterium as a biological treatment can isolate chitosan from FW with a high yield.

2.4. Pectin Extraction from Food Waste

Pectin is another type of polymer that can be naturally found in plant cell walls [39]. It is a heteropolysaccharide important in food, pharmaceutical, and cosmetic industries, [40,41], and is also contained in jams, jellies, and candy. It possesses antidiabetic, antioxidant, anti-inflammatory, antibacterial, and blood cholesterol-regulating properties [42]. Pectin consists of a linear chain of α-(1→4)-linked d-galacturonic acid units [43]. To study for upcycling, Grassino et al. examined several methods of extracting pectin from tomato waste, including conventional extraction with ammonium oxalate/oxalic acid and ultrasound-assisted extraction (UAE) [44]. They concluded that the highest yield of pectin was obtained from conventional extraction at 60 °C while the best quality was yielded using UAE with 15 min of sonification [44]. Shivamathi et al. utilized pineapple peel waste to extract pectin

through UAE and obtained a maximum pectin yield of 16.2% at a 15.2 mL/g liquid-to-solid ratio [45]. Yu et al. used Akebia trifoliata var. australis fruit (akebia quinata) to extract pectin using a citric acid solution that was stirred with the peel powder and filtered [46]. The purified pectin was combined with silver nitrate to form silver nanoparticles, which exhibited strong antibacterial activity with excellent biocompatibility. The authors also used the pectin to make cylindrical porous sponges, called CEPAG1.5. Using *E. coli* and *S. aureus* the authors noticed a clear bacterial inhibition using the CEPAG1.5 sponge. Furthermore, the sponge showed good biocompatibility with human fibroblast skin cells [46]. An in vivo study with rats was carried out on infected wounds using the sponge. Rats with the CEPAG1.5 sponge showed a completely clean and healthy wound healing 6 days after treatment compared to the animals in the control group [46]. Petkowicz and Williams extracted pectin from watermelon rinds by creating alcohol-insoluble residue by boiling the chopped or milled rinds in ethanol, followed by centrifugation [47]. Asgari et al. used the green husks from walnuts to extract pectin by UAE [48]. Demir et al. used lemon peels to extract pectin through acid hydrolysis, which was made into a cryogel with chitosan by dissolving the polymers in acetic acid, which were then cryogelated and freeze-dried for use [49]. Testing of the pectin gel in a glioblastoma cell culture showed that it was non-toxic and supported cell attachment and survival [49]. Saurabh et al. used the UAE method to improve the quality of pectin extracted from jackfruit peel waste [50]. The researchers found that higher quality pectin and yield were obtained using UAE [50]. Lin et al. used chitosan, pectin, and polyvinyl alcohol (PVA) to fabricate electrospun nanofibers that can be used for drug delivery and cell growth [51]. It was discovered that the chitosan–pectin scaffold had a decreased swelling ratio and released the drug more quickly [51]. Additionally, the authors seeded fibroblast cells into the scaffold and observed normal cell morphology, cell proliferation, and similar levels of deposited extracellular type 1 collagen for both scaffolds [51]. Mendez and Lopez extracted pectin from banana peels using hydrochloric and organic citric acids. They noticed a 20% yield with the citric acid compared to the hydrochloric acid, which only had a yield of 7%. In addition, the researchers made polyelectrolyte nanoparticles using the extracted pectin and amphiphilic chitosan and found a good encapsulation efficiency [52]. Pectin can be extracted from waste orange peels with different methods as described elsewhere [53]. For example, highly methoxylated pectin from apple and 5-hydroxytryptophan were conjugated and underwent electron beam irradiation to construct hydrogels that were loaded with tetracycline hydrochloride [54]. The hydrogels showed good biocompatibility and antibacterial properties with different release rates from the porous and nonporous hydrogel. Therefore, the hydrogels would be a good candidate for wound dressing [54]. Frietas et al. detail the different methods for extracting pectin from passion fruit peel, including acid, enzymatic, pressurized, ultrasound-assisted, high hydrostatic pressure, and microwave oven treatments [55]. Govindaraj et al. extracted pectin from jackfruit peels using a toluene and ethanol mixture [56]. Harshith et al. isolated pectin from lemon citrus peels by treatment with hydrochloric acid. The researchers then made a pectin/PVA nanofiber by electrospinning to create a wound dressing in tissue engineering application [57]. One of the more common methods for pectin extraction is UAE, due partly to its environmentally friendly fashion and efficiency [58]. The use of acid and/or ethanol for extraction resulted in a similar yield for pectin extraction. Millian-Linares et al. summarized the extractions of pectin from various wastes with different methods and focused on olives as a potential fuel for pectin extraction [59]. Furthermore, olive pomace is comprised of 35% pectin, similar to other FW such as lemon peel or sugar beet peel, indicating that olive pomace is a good source as well for pectin extraction [59].

2.5. Collagen Extraction from Food Waste

Collagen has numerous industrial values. It is a component in cosmetics, dental composites, and scaffolds for human tissue repair and regeneration [60–65]. It has also been extensively used in biomedical research and clinical applications, including biomanufacturing [66]. Various biomedical applications of collagen have been well summarized

elsewhere [67]. Studies have explored the extraction of collagen from fish waste. Milan et al. used collagen extracted from fresh tilapia skins and mangosteen extract to construct mineralized scaffolds. The collagen was extracted by first removing the fat content, submerging the skin and stabilizing it in a base solution, followed by collagen extraction in acetic acid [68]. Oslan et al. upcycled wasted snapper skin to extract collagens by using various types of acid, such as pepsin, lactic acid, acetic acid, and citric acid [69]. The results showed that the collagen yield was 6.65% using pepsin, followed by 5.79% with acetic acid, 4.15% using citric acid, and 3.19% with lactic acid [69]. Carolo et al. extracted collagen from purple sea urchin waste collected from a restaurant by decellularization of the membrane, and then made a 3D scaffold for wound healing [70]. Hazeen et al. utilized wasted cuttlefish skin to extract collagen and showed a maximum collagen yield of 8.79% under optimal extraction conditions [71]. Abbas et al. pretreated catfish skin with NaOH and then treated it with either acetic acid or pepsin to purify collagen. The authors obtained 86.93% high recovery through the pepsin-soluble collagen method [72]. Martins et al. extracted collagen from the skins of Greenland halibut by using ethanol to remove fat followed by using sodium hydroxide to remove non-collagenous proteins with NaCl and Tris-HCl solution for extraction. However, the maximum yield of type 1 collagen was only 3.8% [73]. Rajbimashhadi et al. highlighted collagen extraction methods from fish industrial wastes and applications of collagen in tissue engineering and wound healing [74]. Since collagen is often extracted from marine life skin, defatting and deproteinization are necessary steps to enhance the quantity and yield of collagen. Decellularization can be an alternative approach for collagen-rich substrates that are non-fatty to produce collagen [75].

2.6. Hydroxyapatite Extraction from Food Waste

Hydroxyapatite is one of the main components of bone tissue. It is composed of calcium, phosphate, and hydroxyl groups to form a lattice structure (Figure 2). It often has a complex crystalline structure of phosphate, hydroxyl, and calcium ions [76]. This material is used in a variety of applications, such as tissue engineering, regenerative medicine, bone substitutes, and drug delivery. Bee et al. explored extracting hydroxyapatite from various types of biowaste [76,77]. These wastes include eggshells, bones, and seafood shells [78]. Borciani et al. focused on marine wastes as a source for extracting hydroxyapatite and made scaffolds for potential in vivo bone tissue engineering applications [79]. Teoh et al. derived hydroxyapatite from waste chicken bones by boiling them in water, deproteinization in acetone and then calcination in a furnace at 1000 °C for 2 h to obtain pure phase hydroxyapatite [80]. Very recently, Boudreau et al. isolated hydroxyapatite from the bones of Atlantic salmon processing waste. They used an enzyme cocktail consisting of two enzymes known as Neutrase and Lipozyme CALB L. The best yield was with 15 μL g^{-1} Neutrase and 7.5 μL g^{-1} Lipozyme CALB L at 40 °C for 6 h of enzymatic hydrolysis [81]. Additionally, nano-sized hydroxyapatite could be isolated from egg shells [82]. The structural formulae of lignin, cellulose, chitosan, pectin, collagen, and hydroxyapatite are exhibited in Figure 2 [8,23,43,83–86].

3. Conversion of Food Waste into Carbon Sources for the Production of Biodegradable Plastics

3.1. Polyhydroxyalkanoate Production from a Variety of Food Waste Using Natural PHA-Accumulating Microorganisms

There are several types of gram-negative and gram-positive bacteria capable of producing biodegradable polymers called polyhydroxyalkanoate (PHA) (Figure 2). PHA polymers have extensive utilities in not only food packaging but also medical products and treatments due to their biocompatibility to human tissues, biodegradability, and physiochemical properties. Hence, PHA is a material commonly used in regenerative medicine, tissue engineering, and various medical devices [87]. Particularly, a short-chain length PHA, polyhydroxybutyrate (PHB), is readily accumulated by several types of bacteria. *Curpriavidus necator* (*C. necator*) H16, also known as *Ralstonia eutropha*, *Alcaligenes eutrophus*, *Hydrogenomonas eutropha*, and *Wautersia eutropha* in the literature, is often used for PHA

production. Various types of FW, including but not limited to spent coffee grounds, rice straw, rice husks, and waste cooking oil, have been studied for their potential use as raw materials for PHA production (Figure 1C). Table 1 summarizes diverse types of FW used for biodegradable polymer production by means of PHA-accumulating microorganisms. Spent coffee grounds contain 11~20 wt% of coffee oil that can be extracted using solvent n-hexane [88]. Researchers have utilized the coffee oil extracted as a carbon source at 30 g/L to culture *C. necator* H16 in a synthetic medium containing mineral salts, resulting in a high production of 55.4 g dry cell weight (DCW)/L, 89.1% PHB content in biomass, and 1.33 g/L/h volumetric productivity (Table 1) [89]. In another study, oil extracted from spent coffee grounds through supercritical extraction was used for producing PHA in *C. necator* [90]. The culture obtained 16.7 g DCW/L with a polymer content of 78.4% [90]. Researchers also examined the potential to use rice straw hydrolysates as a carbon source to produce PHA with *C. necator.* They optimized the ratio of carbon to nitrogen sources (C/N ratio) and found that PHA accumulation increased under higher degrees of nitrogen deficient conditions of 0.01 g/L of NH_4Cl and a C/N ratio of 160:1 had a PHA content of 21% [91]. Furthermore, rice husks were pretreated using potassium hydroxide and enzymatic hydrolysis to obtain sugars from the waste. The pretreatment was able to convert the rice husks to approximately 20 g/L total sugars [92]. Using the sugar-containing hydrolysate as the carbon source in PHA bacterial cultures, it was found that *B. cepacia* consumed rice husks-derived sugars more efficiently than *C. necator,* resulting in 7.8 g DCW/L and 50% PHA content [92].

Several other types of oil wastes have been studied for biodegradable polymer production via *C. necator* fermentation. Wastewater generated from palm oil milling called palm oil mill effluent (POME) was treated by an anion exchange resin separation column to enrich acetic acid concentration from the waste [93]. The concentrated acetic acid was used as a sole carbon source in the fed-batch production of PHA by *Alcaligenes eutrophus*. Within 17 h of the culture, the overall volumetric productivity of PHA was approximately 0.09 g PHA/L/h and PHA content of 45% [93]. In another study, Kamilah et al. collected palm oil-derived cooking oil waste from households and eateries and utilized the oil waste as a carbon source for the cultivation of PHB-accumulating bacteria. It was noted that there is a higher PHB yield using leftover cooking oil compared to fresh cooking oil [94]. As cooking oil is rich in fatty acids, when cooking oil waste was supplemented in a mineral medium at 20 g/L for *C. necator* DSM 428 culture, 10.4 g DCW/L and PHB concentration of 3.8 g/L were achieved, resulting in 37% PHB content [95]. Similarly, using sunflower oil waste collected from households as a sole carbon source, the same bacteria grew to 11.4 g DCW/L and a PHB concentration of 5.8 g/L, increasing PHB content to 57% (Table 1) [95]. Sandhya and Kanmani utilized paddy straw mushrooms as the carbon source for producing PHA using *C. necator* and achieved 37.6% PHA accumulation at 19.2 g DCW/L [96]. Obruca et al. studied wasted rapeseed oil to produce PHA in *C. necator* fermentation. By adding propanol at 1% (*v*/*v*) during the fed-batch culture, they obtained 138 g biomass/L and 105 g/L PHA, demonstrating an effective PHA production using propanol as a precursor of polymer biosynthesis together with waste rapeseed oil [97].

Additional FWs have been investigated for converting to PHA and some types of FW have demonstrated a promising conversion efficacy. For instance, Haas et al. saccharified waste potato starch through thermal and enzymatic processes to obtain highly concentrated glucose. The saccharified solution was used as a carbon source in *Ralstonia eutropha* NCIMB 11599 fed-batch culture. The authors achieved 179 g/L biomass, 94 g/L PHB with 52.5% PHB content, and a productivity of 1.47 g/L/h (Table 1) [98]. Salgaonkar and Braganca studied four halophilic archeal isolates to utilize sugarcane bagasse as a carbon source for the biosynthesis of PHA. The authors reported the maximum PHA content of 50.4% from *Halogeomatricum borinquense* E3 at a 4.15 g DCW/L [99]. Cesario et al. utilized *Burkholderia saccharum* in fed-batch cultures with wheat straw hydrolysates. A productivity of 0.7 g/L/h poly(3-hydroxybutyrate-co-4-hydroxybutyrate) P(3HB-co-4HB) was obtained [100]. Cheese whey is a liquid byproduct from the manufacture of cheese. Colombo et al. compared

the PHA production using two different fermented cheese whey in a preselected mixed microbial culture. They found that a high PHA production was from the whey that had added propionic and valeric acids in the culture medium, leading to the production of 70 g PHA per kg of cheese whey. As propionic and valeric acids were part of the substrates in the culture, the polymer accumulated composed of 40% of 3-hydroxyvalerate (HV) and 60% of HB 3-hydroxybutyrate (HB) [101].

FW conversion into biodegradable polymers through fermentation of different bacterial stains has also been widely investigated. Interestingly, Follonier et al. used a two-step fermentation with the hydrolyzed fruit pomace as a bacterial culture substrate and waste frying oil as a PHA precursor in *Pseudomonas resinovorans* fermentation. They investigated nine different types of FW, including apricots, cherries, grapes, and waste frying oil, for their utility in producing PHA [102]. A maximum production of 21.3 g PHA per liter of pomace was observed with the cherry pomace [102]. Kourmentza et al. investigated the use of sunflower-derived cooking oil as a substrate for PHA production with *Burkholderia thailandensis* E264 strain. They cultured the stain in a nutrient medium containing peptone and meat extract and obtained 12.2 g DCW/L and a PHA concentration of 7.5 g/L (Table 1) [103]. In another study, spent coffee grounds were thermal and enzymatically treated to break up cellulose for releasing saccharides in the waste. The resultant solution, called hydrolysate of SCG (SCGH), was rich in sugar, containing approximately 50 g/L of total sugars, 23.6 g/L of mannose, 17.3 g/L of galactose, 3.9 g/L of glucose, 2.8 g/L of arabinose, and 2.7 g/L of cellobiose [104]. Using the SCGH as the carbon source in *Burkholderia cepacia* culture, a maximum of 4.9 g DCW/L and 54.8% of PHA content were obtained [104]. Kachrimanidou et al. used sunflower seed with *C. necator* to produce poly(3-hydroxybutyrate-co-3-hydroxyvalerate) (PHBV). Uniquely, the fungal strain *Aspergillus oryzae* was used to lyse *C. necator* to recover PHB [105]. The optimum temperature and an uncontrolled pH resulted in a recovery of 98% and 96.7% of PHBV and P(3HB-co-12 mol% 3HV), respectively [105]. Sawant et al. utilized corn to accumulate PHA in *Paracoccus* sp. *LL1* culture. The corn was first hydrolyzed with a cellulase cocktail and then fed to the bacteria. The authors reported that there was a 13.41 g DCW/L and PHA content of 72.4% [106]. Sindhu et al. utilized three different bacterial strains, i.e., *B. firmus NII 0830*, *Bacillus sphaericus NII 0838*, and *Paracoccus denitrificans*, to compare their efficiency in producing PHB using rice straw-derived pentose sugar hydrolysate as a carbon source. They noted that the *Bacillus firmus NII0830* showed a maximum 1.9 g DCW/L and PHB concentration at 1.7 g/L, leading to a PHB cell content of 89% [107].

Other bacterial stains have been studied for PHA production from a variety of FW types. Cesario et al. prepared lignocellulosic hydrolysates from wheat straw and used as a carbon source to culture *Burkholderia sacchari*. The work observed 70% PHB content per DCW with a PHB concentration of 3.8 g/L [108]. Kulpreecha et al. utilized a newly isolated *Bacillus megaterium* strain to make PHB using sugarcane molasses and urea as a carbon and nitrogen source, respectively. Under optimized conditions, the authors obtained a PHB content of 42% per DCW and 1.27 g/L/h PHB productivity [109]. In another study, liquid bean curd waste together with sucrose were used as carbon sources for producing PHA in *Alcaligenes latus* and received 2.48 g/L of PHA with 66.6% content (Table 1) [110]. Lopes et al. reused hydrolyzed sugarcane bagasse to feed *Burkholderia* sp. *F24* and produced 25.04 g DCW/L with a PHB content of 49% [111]. Pais et al. recycled cheese whey as the substrate to produce HBHV in *Haloferax mediterranei* culture. They reported a polymer content of 53% and productivity of 4.04 g/L/day [112]. Patel et al. added a hydrolyzed pea shell slurry to a defined culture medium and examined several bacterial strains, including *B. cereus EGU3*, *EGU43*, *EGU44* strains and *B. thuringiensis EGU45* for the PHA accumulating capacity. However, the best batch culture led to a PHB production of only 1.69 g/L in *Bacillus cereus* EGU44 [113].

Table 1. Summary of different types of food waste used for biodegradable polymer production by means of PHA-accumulating microorganisms.

Food Waste	Bacterium	DCW * (g/L)	PHA/PHB ** Content (%)	Reference
Spent coffee grounds	*Curpriavidus necator*	55.4	89.1	[89]
Spent coffee grounds	*Cupriavidus necator* DSM 428	16.7	78.4	[90]
Saccharified spent coffee grounds	*Burkholderia cepacia* CCM 2656 (ATCC 17759)	4.9	54.8	[104]
Rice straw	*Bacillus firmus* NII 0830	1.9	89	[107]
Rice husk	*B. cepacia USM* (JCM 15050)	7.8	50	[92]
Waste cooking oil	*Cupriavidus necator* H16	15.5	70	[94]
Used cooking oil	*Cupriavidus necator* DSM 428	10.4	37	[95]
Used sunflower oil	*Cupriavidus necator* DSM 428	11.4	57	[95]
Waste rapeseed oil with 1% of propanol	*Cupriavidus necator* H16	138	75	[97]
Sunflower-derived cooking oil	*Burkholderia thailandensis* E264	12.2	61	[103]
Paddy straw mushrooms	*Ralstonia eutropha* MTCC 1472	19.2	37.6	[96]
Sugarcane bagasse	*Halogeometricum borinquense* E3	4.15	50.4	[99]
Sugarcane bagasse	*Burkholderia* sp. F24	25.04	49	[111]
Saccharified waste potato starch	*Ralstonia eutropha* NCIMB 11599	179.0	52.5	[98]
Cheese whey	*H. mediterranei* (ATCC 33500)	16.01	53	[112]
Corn stover	*Paracoccus* sp. *LL1*	13.41	72.4	[106]
Sugarcane molasses	*Bacillus megaterium* BA-019	72.6	42	[109]
Liquid bean curd with initial sucrose at 25 g/L	*Alcaligenus latus*	3.73	66.56	[110]

* DCW: dry cell weight. ** PHA/PHB: polyhydroxyalkanoate/polyhydroxybutyrate.

3.2. Polyhydroxyalkanoate Production from Food Waste Using Recombinant Strains

With modern DNA cloning technology and metabolic flux network analysis, researchers continue to optimize conditions for PHA production by means of genetic modification of the fast-growing bacterium, but naturally cannot accumulate polymers. Recombinant *E. coli* strains with added genes and/or pathways for PHA production have been constructed. Table 2 summarizes FW as substrates for biodegradable polymer production by means of recombinant bacterial strains. Hong et al. examined the recycling use of soy waste for PHB accumulation in a recombinant *E. coli* strain, where a *phb* operon was carried by a plasmid DNA. The authors reported a PHB content of 27.8% with the recombinant *E. coli* [114]. Furthermore, the authors reported that the lac promoter was the most efficient among the three promoters investigated (lac promoter, T7 promoter, and the normal σ70 promoter) [114]. Law et al. utilized *E. coli* and a plasmid to introduce PHA-producing genes into the cells and to produce PHA from malt and soy wastes. They found that the *E. coli* HMS174 strain showed the highest yield in the production of PHBV with 10.27 g DCW/L and a PHA content of 43% [115]. An *E. coli* strain harboring the genes from *C. necator* for PHA production with whey as a carbon source was able to produce PHB at a concentration of 5.2 g/L with 81% PHB content [116]. In another study, a recombinant *E. coli* strain CML3-1 harboring *C. necator* PHB-synthesis genes, *phbC*, *phbA* and *phbB* was constructed [117]. There was a PHB concentration of 7.83 g/L with a PHB content of 21.73% by this strain when cheese whey was used as a carbon source, although the *E. coli* reached 36 g DCW/L [117]. Another recombinant *E. coli* CGSC 4401 harboring *Alcaligenes latus* PHA biosynthesis genes was created. When the strain was cultured in a flask, the biomass and PHB concentrations were 6.6 g/L and 5.0 g/L, respectively [118]. An engineered strain of *C. necator* overexpressing (R)-specific enoyl coenzyme-A hydratase (phaJ) and PHA synthetase (phaC2) with deletion of acetoacetyl Co-A reductases (phaB1, phaB2, and phaB3)

has been employed for producing PHA using extracted oil from coffee waste oil as a carbon source. The study produced a polymer poly(3-hydroxybutyrate-co-3-hydroxyhexanoate) (P(HB-co-HHx)) at PHA content of 69%. However, the biomass concentration was only 0.89 g/L [119]. These studies demonstrated the values of distinct FW types for the production of biodegradable plastics. Reuse of FW rather than FW landfill possesses great potential not only to significantly reduce the emissions of greenhouse gases from landfilled FW but also to reduce the cost of current PHA production in industry, where expensive refined sugars such as glucose and fructose are used.

Table 2. Summary of food waste as substrates for biodegradable polymer production by means of recombinant bacterial strains.

Bacterium	Inserted Genes	Food Waste	DCW * (g/L)	PHA/PHB ** Content (%)	Reference
E. coli	*phb* operon	Soy waste	3.03	27.83	[114]
E. coli	*phaC*, *phaA*, and *phaB*	Malt waste	10.27	43	[115]
E. coli	*Alcaligenes eutrophus polyhydroxyalkanoate (PHA) biosynthesis genes*	Whey	6.42	81	[116]
E. coli	*C. necator phbC*, *phbA* and *phbB genes*	Cheese Whey	36	21.73	[117]
E. coli	*A. latus PHA biosynthesis genes*	Whey	6.6	76	[118]
E. coli	*Amp*, *phaC1 gene* from *P. aeruginosa* and *PHA operon* from *R. eutropha*	Corn starch and soybean Oil	0.92	5.9	[120]
Cupriavidus necator	*Overexpressing phaJ* and *PHA synthetase (phaC2)* with deletion of *phaB1*, *phaB2*, and *phaB3*	Coffee waste oil	0.89	69	[119]

* DCW: dry cell weight. ** PHA/PHB: polyhydroxyalkanoate/polyhydroxybutyrate.

4. Diverse Types of Food Waste-Derived Materials as Essential Components for Fabricating Medical Products

Materials made from various types of FW described above can be useful in creating medical products, which are highlighted in Figure 3. Different kinds of hydrogels containing FW-derived components have shown their potential usage in medical treatments. These products include hydrogels as wound dressings and nanoparticles for drug delivery. The most promising discoveries are those that have been tested in animal models. Figure 1B highlighted FW-derived novel biomaterial combinations used to construct functional hydrogels for medical treatments. Pandit et al. created a spongy wound dressing by combining carboxymethyl tamarind seed polysaccharide, pectin, and glycerin with a layer of moxifloxican-filled alginate beads in the middle [121]. The formulated wound dressing showed high wound fluid adsorption, good endurance, and high drug release. In wounded rats, the dressing showed almost complete closure in 17 days [121]. Interestingly, pectin has also been investigated in combination with papain to create a spray on topical gel to help skin wound healing. This material displayed a better healing progression compared to controls in a rabbit experiment [122]. Lignin extracted from coconut husk has been incorporated in a hydrogel including polyethylene glycol, polypropylene glycol (PPG), and polydimethylsiloxane together with 1% vitamin C, (+)-catechin hydrate, and Trolox [15]. Upon applying it to mice with burned skin, the study demonstrated that the lignin-containing hydrogel processed strong antioxidant activity and effectively promoted wound recovery compared to a slower recovery rate in the mice without lignin addition [15]. Florentini et al. used a combination of zein, pectin, and soy lecithin with vitamin C to construct a microfibrous scaffold to test its potential healing properties. The fibers were made by electrospinning zein and pectin, which was crosslinked with Ca^{2+} to create a hydrogel [123]. In vivo testing on skin UVB-burned models showed reduced inflammatory cytokines in the injured area [123]. In a study by Cui et al,. an organohydrogel as an antifreeze wound dressing was formulated by using the aforementioned durian rind

cellulose powder and a LiOH/Urea mixture initially (Figure 1B) [25]. To obtain the final antifreeze wound dressing, the initial hydrogel was first submerged in glycerol to make an organohydrogel and then in a yeast phenolic stock solution to obtain its antimicrobial properties. The durian rind cellulose organohydrogel was applied as a wound dressing on wounds in pig skin for the experimental group and moist gauze was used as the control group. Their results suggested low microbial development, which could be resolved by periodically replacing the dressing. The study indicated that the developed organohydrogel has the potential to act as a practical wound dressing [25].

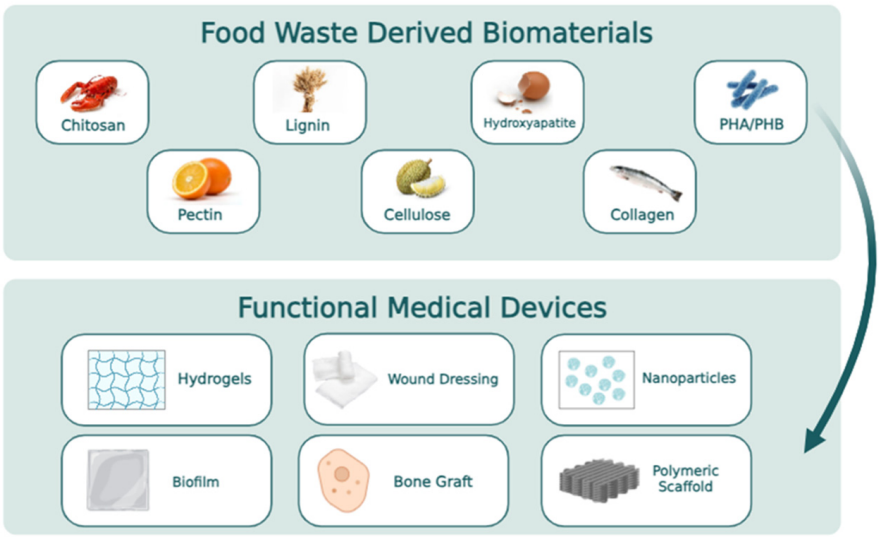

Figure 3. Food waste-derived materials are essential components to fabricate medical devices for diverse applications in tissue engineering and drug delivery.

In addition, lignin extracted from wheat straw has been combined with alginate to create a hydrogel [16]. Mouse fibroblast cells cultured on the hydrogel showed good cell adhesion with little or no cytotoxicity [16]. Amirian et al. combined amidated pectin and oxidized chitosan to fabricate crosslinked hydrogels and showed good swelling capacity and biocompatibility [124]. Furthermore, Kocaaga et al. utilized low-methoxyl pectin and zeolite to make crosslinked hydrogels that can hold drugs for controlled release during wound healing. They found the hydrogels are non-cytotoxic, support cell proliferation and migration with good swelling properties [125]. Carolo et al. made a marine collagen scaffold by adding the collagen at 2 g/L with 0.01% TritonX-100 to rubber silicon molds, frozen overnight, and lyophilized overnight [70]. To test the scaffold the researchers made a wound in adult male rats with a section of the dorsal column cut out and placed the scaffold to the wound area. Compared to a commercially available brand for dermal regeneration, the marine collagen scaffold showed similar biocompatibility, stimulated angiogenesis, and the deposition of mature collagen [70].

Furthermore, FW-derived materials have been explored to fabricate nanoparticles for drug delivery (Figure 3). Shahzad et al. used chitosan from crab shells and sodium alginate to fabricate cefazolin-encapsulated chitosan nanoparticles [126]. The resultant nanoparticles were then loaded onto a film containing sodium alginate and pectin and crosslinked using calcium chloride. Their experimental results suggested that there was an equal distribution of the drug cefazolin in all parts or areas of the film. The film allowed fluid uptake and drug release, no rapid swelling, and was antimicrobial [126]. In another study, lignin silica nanohybrid particles were created and exposed to human bone marrow derived mesenchymal stem cells to examine its impact on cell viability and cell morphology. The novel materials showed good cytocompatibility and successful osteogenic differentiation from the stem cells [13]. Moreover, lignin extracted from wheat straw was used to synthesize silver nanoparticles by mixing a lignin solution with a silver nitrate

solution. The anticancer activity of the nanoparticles was investigated using the SKOV3 cancer cell line. It was confirmed that the lignin-containing nanoparticles are capable of declining the cell viability in a dose-reliant mode [14]. Dai et al. used alkali lignin extracted from corn cob through a hydrothermal treatment to fabricate nanoparticles with an organic solvent solution. The nanoparticles were then assembled with resveratrol and magnetite to create a nanodrug carrier. The materials exhibited good stability, biocompatibility, and a relatively high drug loading capacity of over 20% wt. In addition, the materials inhibited tumor growth and improved experimental animal survival rates [18].

Additionally, the components extracted from varied FW types have been incorporated into other materials to fabricate film, nanoparticle, and powder for medical treatments (Figure 3). Koshy et al. constructed a biofilm containing pectin from citrus peels and *hemigraphis alternata* extract as a potential anticancer wound care therapy [127]. Kamel et al. incorporated banana peel powder into a chitosan matrix and showed good antimicrobial properties against gram negative/gram positive and yeast bacteria [128]. Li et al. used eggshell membrane powder and chitosan to create a film. The material showed good wound healing and swelling properties, and antibacterial activity, suggesting its potential as a wound dressing [129]. Govindaraj et al. combined pectin from jackfruit peel with $CaCl_2$ and then K_2HPO_4 to yield fine bionanocomposite particles. These nanoparticles showed good biocompatibility that signifies its potential application as a bone graft material [56]. In another study, researchers have incorporated citrus pectin into a copper-based metal–organic framework with folic acid. The resultant fibers possessed antibacterial activity and were biocompatible with good tensile strength [130]. Moreover, FW-derived lignin was blended with chitosan to make a composite. The antibacterial activity of the composite was investigated with the zone inhibition method using *S. aureus* and *E. coli* and cell viability was tested in mouse embryonic fibroblast culture [17]. The composite showed strong antibacterial properties and biocompatibility to the cells, indicating that it can be successfully used as a wound dressing [17].

Collagen has been an excellent biomaterial in scaffolding to support human cell attachment, proliferation, and differentiation [61,63,131]. Irastorza et al. detailed the use of collagen and chitosan as materials for tissue engineering, including the preparation techniques, properties, and applications of various materials such as films, sponges, hydrogels, and fibers [132]. Milan et al. made a mineralized scaffold using collagen derived from tilapia skin and mangosteen extract. The study indicated the potential of this material to facilitate the formation of bone tissue, due to the stability of the collagen chains and high porous structure that allow for calcium phosphate nucleation [68]. Arslan et al. combined keratin from human hair, collagen from jellyfish, and hydroxyapatite from eggshells to form a scaffold for bone tissue engineering [82]. After crosslinking and freeze drying the scaffold, the authors seeded human adipose mesenchymal cells to the scaffold and observed an osteogenic effect on the cells, as there was a calcified matrix and increased expression levels of osteoporin and osteonectin [82].

PHAs are extensively used to make biodegradable medical products. Kalia et al. summarized the different types of biodegradable implants with applications in tissue engineering, wound dressings, and drug delivery [133]. Bonarstev et al. detailed the various types of PHA and their usages in commercially available medical products [87]. Najah et al. created a biocomposite containing calcium phosphate, PHA, and chitosan to enhance the strength of the composite for bone graft applications [134]. The study showed that maximum tensile stress and elastic modulus could be achieved at 15 wt% of calcium phosphate [134]. Injorhor et al. fabricated electrospun fibers from PHA, biodegradable polylactic acid, and nano-hydroxyapatite from fish scales. The material showed tensile strength, thermal stability, and is in vitro degradable, which implies its potential use in bone tissue engineering [135]. Phuegyod et al. made a porous scaffold using polymer PHBV with 50% HV content biosynthesized from *C. necator H16* and tested the polymer's utility for periodontal tissue engineering [136]. After seeding human gingival fibroblasts and periodontal ligament stem cells to the polymer scaffolds, high proliferation rates,

good cell adhesion, and good morphology were observed, indicating the potential for periodontal tissue engineering [136]. Ghadirian et al. fabricated a PHB electrospun scaffold with the addition of halloysite nanotube, a naturally occurring tubular clay material, and investigated the scaffold application for cartilage tissue engineering [137]. It was found that the scaffold possessed better mechanical properties, resistance to degradation, and supported proliferation of chondrocytes on the scaffold [137]. Other types of polymers were developed to make medical devices. For instance, Song et al. fabricated swellable polymer with branched L-borneol antibacterial agents. It exhibited similar antibacterial performance as chitosan and other natural materials [138]. In addition, hyper-crosslinked polymers have similar properties to those of natural materials, are valuable in drug delivery and possess antibacterial properties. They are considered a promising new class of materials for biomedical applications [139].

5. Conclusions

It is estimated that one-third of the world's food is wasted annually with growing economic and population growth. FW is one of the major sources attributing to environmental pollution and global climate change due to the emissions of greenhouse gases to the atmosphere from landfilled FW. Extensive efforts have been made to explore the recycling use of a variety of FW sources ranging from plant FW to marine waste for improving environmental sustainability. Turning FW into functional biomaterials and medical products enables FW streams to be upcycled for manufacturing value-added products, instead of contributing to greenhouse gas emissions and environmental pollution. Although converting FW into diverse value-added materials is a promising approach for improving environmental protection and has been extensively investigated, significant challenges exist. For instance, most of the studies have focused on isolating a material from a single type of FW on a small scale. A material's isolation process may encompass chemical, physical, biological or a combination of them. Hence, the scale-up study is essential to optimize extraction efficacy and reduce cost on a large scale to attract the development of a new industry for the sustainability of FW reuse. The optimization includes development of green isolation method in case a toxic solvent is used for isolation. Furthermore, knowledge of the extraction efficacy of the materials from a mixture of varied FW types would offer insights into the commercialization of FW recycling technologies. This is critical from an industrial point of view, as many FWs could be constantly collected and transported to repurposed processes from food processing industries and kitchen wastes in restaurants. To overcome these bottlenecks, it is necessary to draw attention from governments, industries, and researchers and for these three parties work jointly for the realization of industrial-scale manufacturing of products through FW upcycling.

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