7th International Food Operations & Processing Simulation Workshop
18th International Multidisciplinary Modeling & Simulation Multiconference

ISSN 2724-0355 ISBN 978-88-85741-69-0 © 2021 The Authors.
doi: 10.46354/i3m.2021.foodops.005

Agricultural waste as a source of innovative and compostable composite biopolymers for food packaging: a scientific review

Anna Ronzano[1], Roberta Stefanini[2], Giulia Borghesi[1], Giuseppe Vignali[2]
[1] Cipack Centre, University of Parma, Parco Area delle Scienze, 43124 Parma (Italy)
[2] Department of Engineering and Architecture, University of Parma, Parco Area delle Scienze, 181/A, 43124 Parma (Italy)
*Corresponding author: Giuseppe Vignali. Email address: giuseppe.vignali@unipr.it

Abstract

The recovery of agriculture waste is one of the challenges of 2030 Agenda. Food and Agriculture Organization states that 30 % of the world's agricultural land is used to produce food that is later lost or wasted, and the global carbon footprint corresponds to 7% of total greenhouse gases emissions. Alternatively, natural fibers contained in food and agricultural waste could be a valuable feedstock to reinforce composite biopolymers contributing to increase mechanical properties. In addition, the use of biopolymers matrix could contribute significantly to reduce the environmental footprint of the biobased compounds.

Based on these premises, a regional project in Emilia-Romagna, aims to enhance agricultural waste to produce food packaging materials which in turn would contribute to the reduction of green raw materials used. This article reviews the state of art of composite biopolymers added with fillers extracted by food and agricultural waste, analyzing the literature published on scientific databases such as Scopus. The characteristics, advantages and drawbacks of each innovative sustainable material will be studied, trying to compare their various properties.

The results of the work could guide companies in the choice of eco-sustainable packaging and lay the foundations for the development of the mentioned regional project.

Keywords: food packaging; composite biopolymers; agricultural waste; food waste; natural fibers

1. Introduction

The global bioplastics production capacity is set to increase up to 2.8 million tonnes in 2025 and 47% of the total bioplastics market in 2020 is the field of packaging (European bioplastics, 2020). The improvement of physical and mechanical properties, such as the growing attention for a lower environmental impact of packaging, is allowing the growth of research and investment on these promising alternative materials. Food and Agriculture Organization states that 30 % of the world's agricultural land is used to produce food that is later lost or wasted, and the growing population will result in a boost of demand for agricultural products up to 50 % until 2050, that will be related to an increase of the wastes related to agricultural and food sector (FAO Food and Agricultural Organization of the United Nations, s.d.). From this point, biobased plastics and value-added compounds derived from several wastes are promising alternative materials potentially applicable in packaging sector with a lower environmental footprint. The aim of this review is to investigate the recent publications on composite food packaging blended with fillers extracted by renewable resources, in order to understand how several packaging properties are influenced. This area of research is useful to the related regional project in Emilia Romagna, which purpose is the realization of low-cost bio-composite material with potentially application in food packaging.

2. Material and methods

2.1. Research methodology

A literature review was carried out to investigate the state of the art of the composite materials applied in food packaging, considering several types of waste and focusing on polymers extracted by renewable resources. A computerized research was made using Scopus database (Elsevier, 2021), looking for terms in "title, abstract, keywords", including only "reviews", "articles" and "Conference paper" written in English, at the final publication stage, published all over the world from 2017. The research methodology is reported in Figure 1.

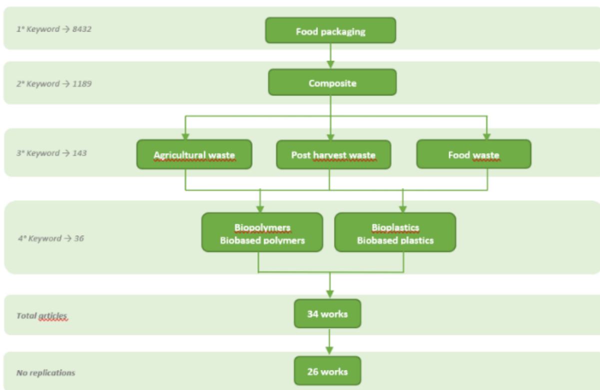

Figure 1. Research methodology used on Scopus database, using different keywords, finding articles, reviews ad conference paper published since 2017

Firstly, a search was conducted using the keyword "Food packaging" (f.p.) in order to get an overview of the papers referred to this field and narrow the research to the category of "composite" materials (c), by adding the second keyword. Starting to the samples of 8432 publications referred to food packaging, 1189 articles report to category "composite". Adding "compostable" as a keyword to composite food packaging (c. f. p.), the search took only to 12 articles, and it was excluded due to the low obtained results.

Then, the analysis was focused on the several types of waste connected to the food and agricultural sector with the words "food waste", "agricultural waste" and "postharvest waste". The results of this second step are shown in Figure 2 and Figure 3.

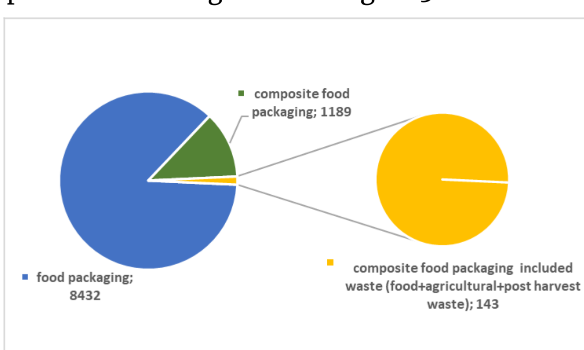

Figure 2. First literature analysis results: number of publications

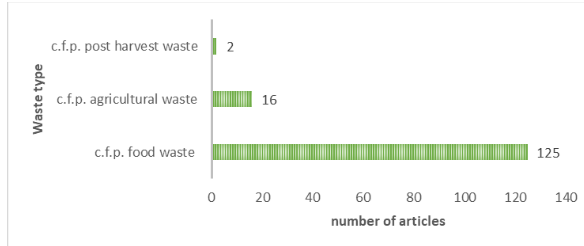

Figure 3. Number of publications of composite food packaging (c.f.p.) for waste type

Consequently, in order to define the research connected with the regional Project, the search mentioned above was combined with the keywords referred to the material "biopolymers", "biobased polymers", "bioplastics", "biobased plastics": 34 publications were found only for food and agricultural waste. Although no work was found with the combination of material with postharvest waste, we also considered the 2 articles in the earlier stage, which are useful for this topic.

Finally, reading carefully the authors name and titles the replications were removed, starting from 34 samples, a total of 26 works were considered for the review analysis, as shown in Figure 4.

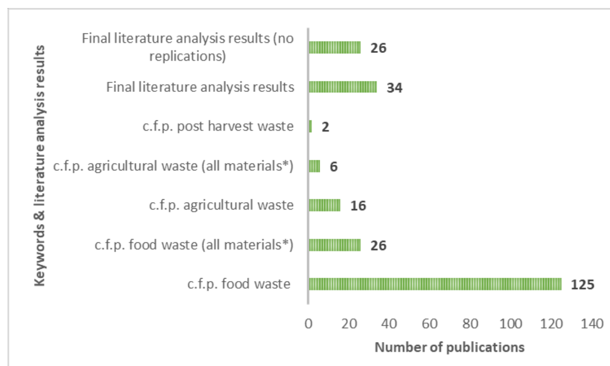

Figure 4. Final literature analysis results

3. Results

3.1. Literature overview

Starting the literature analysis, it can be noticed that composite food packaging related to food, agricultural and postharvest waste is a very restricted field of research: 26 papers were found. Figure 5 shows that literature analysis took to 16 articles and 10 reviews, no reference papers were obtained. Even though the publications are less than or equal to seven articles between 2017-2020, six papers were published in July 2021, and we can suppose that the number will increase until the end of the year, as can be expected by the overview of publications over the years reported in Figure 6. This result underlines that this area of research is took attention on scientists and researchers that are searching for alternative renewable resources in the field of composite food packaging.

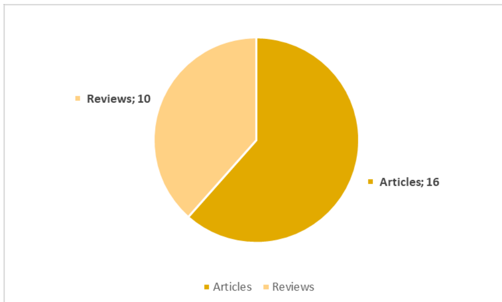

Figure 5. Articles and reviews about composite food packaging (food +agricultural+ postharvest waste) all materials

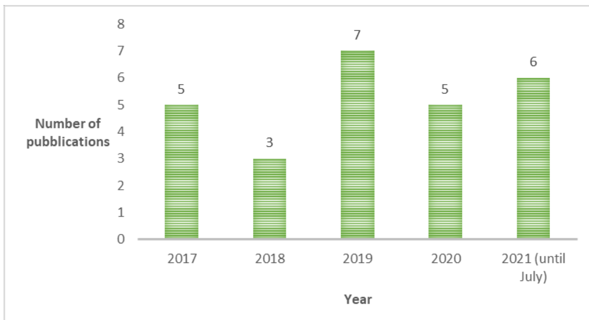

Figure 6. Number of publications over years

The literature analysis has shown that scientific publications on c. f. p. combined with food waste included 15 articles, greater compared to the combination with agricultural waste that resulted to 4 works. In addition, as reported in Figure 7, both combination keywords for c. f. p. food and agricultural waste, have shown several results based on multiple definition of materials derived from renewable resources: biopolymers, biobased polymers, bioplastics and biobased plastics. Main contents are presented in the next paragraph of discussion.

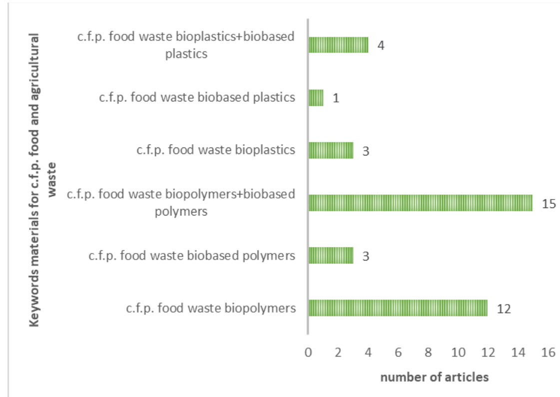

Figure 7. Number of articles for different materials and several waste type

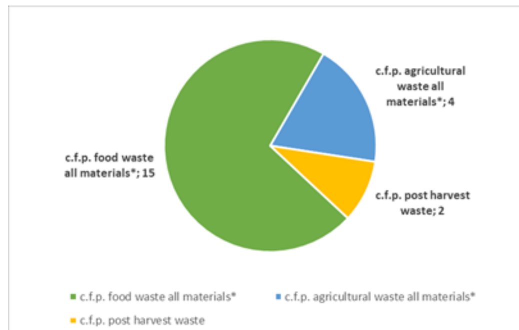

Figure 8. Number of publications of c. f. p. related to the several waste type

Considering all these materials, in Figure 8 it can be noticed that the higher number of papers is referred to food waste compared to agricultural and postharvest waste.

Finally, Table 1 summarizes the found articles in the current literature review, considered respectively research keywords, authors and year of publication, title, and authors keywords. With the purpose of a clear comprehension, the results of c. f. p. were divided on the different polymer matrix, classified into two macro categories: bioplastics/ biobased plastics that includes all the materials according to European Bioplastics definition and biopolymers/ biobased polymers which considers all the remained alternative polymers derived from renewable sources polymers (e. g. cellulose, hemicellulose, starch & lignin, legumes, chitosan). In the end, the blending of each category of polymers with the fillers extracted by the different waste type mentioned above, was discussed.

Table 1. List of the 26 publications studied in the literature review analysis

Research keywords	Authors	Year of publication	Title	Author Keywords
Composite food packaging biopolymers food waste	Udayakumar, et al.	2021	Biopolymers and composites: Properties, characterization and their applications in food, medical and pharmaceutical industries	Biopolymer; Characterization; Drug delivery; Food packaging; Scaffold; Tissue engineering
	Boey, et al.	2021	A review of the applications and biodegradation of polyhydroxyalkanoates and poly (Lactic acid) and its composites	Biodegradation; PHA-based composites; PLA-based composites; Poly (lactic acid); Polyhydroxyalkanoates
	Kharrat, et al.	2020	Minimally processed date palm (Phoenix dactylifera L.) leaves as natural fillers and processing aids in poly (lactic acid) composites designed for the extrusion film blowing of thin packages	Biocomposites; Biopolymers; Film blowing; Palm leaf fillers; PLA
	Ortiz-Barajas, et al.	2020	Torrefaction of coffee husk flour for the development of injection-Molded green composite pieces of polylactide with high sustainability	Circular Bioeconomy; Coffee husk; Green composites; PLA; Torrefaction; Waste valorization
	Salwa, et al.	2019	Green bio composites for food packaging	Bio-based packaging; Biodegradable packaging; Green biocomposites; Sustainable packaging
	Jamróz, et al.	2019	The effect of nanofillers on the functional properties of biopolymer-based films: A review	Antimicrobial activity; Biopolymer films; Film mechanical properties; Film permeability; Food packaging systems; Functional properties; Nanocomposite materials; Nanofillers
	Salwa, et al.	2019	Analytic Hierarchy Process (AHP)-based materials selection system for natural fiber as reinforcement in biopolymer composites for food packaging	AHP; AHP rating mode; Design process; Food packaging; Green biocomposites; Natural fiber selection
	Benito-González, et al.	2018	Potential of lignocellulosic fractions from Posidonia oceanica to improve barrier and mechanical properties of bio-based packaging materials	Biopolymers; Food packaging; Lignocellulosic fractions; Posidonia oceanica; Starch
	Otoni, et al.	2018	Optimized and scaled-up production of cellulose-reinforced biodegradable composite films made up of carrot processing waste	Biocomposite; Biodegradability; Biopolymer; Cellulose derivative; Daucus carota L.; Residue
	Borah, et al.	2017	Ultrasound treated potato peel and sweet lime pomace based biopolymer film development	Biopolymer film; Bread; Clove essential oil; Potato peel (PP); Sweet lime pomace (SLP); Ultrasound
	Nandhavathy, et al.	2017	Optimization of pomegranate peel fibers reinforced with polyvinyl alcohol biocomposite film using response surface methodology	Biocomposite film; Pectin; Polyvinyl alcohol; Pomegranate peel; Response surface methodology
	Attaran, et al.	2017	Materials for food packaging applications based on bio-based polymer nanocomposites	bio-based polymer; biodegradable; Food packaging; layered silicates; nanocomposite
Composite food packaging biobased polymers food waste	Roso, et al.	2021	Experimental methods in chemical engineering: Barrier properties	barrier properties; gas permeability; nanoparticles; polylactic acid; polymer
	Wu, et al.	2019	Antibacterial Properties of Biobased Polyester Composites Achieved through Modification with a Thermally Treated Waste Scallop Shell	antibacterial activity; biocompatibility; biodegradation; polymer-matrix composites; waste white scallop shell
	Cinelli, et al.	2019	Biocomposites based on polyhydroxyalkanoates and natural fibres from renewable byproducts	Biocomposites; Food by-products; Mechanical properties; Polyhydroxyalkanoates; Pukanzsky's model
Composite food packaging bioplastics food waste	Diaz, et al.	2020	Thermoformed containers based on starch and starch/coffee waste biochar composites	Biochar; Biodegradation; Bioplastics; Coffee waste; Polycaprolactone; Starch
	Marzuki, et al.	2020	The effect of jackfruit skin powder and fiber bleaching treatment in pla composites with incorporation of thymol	Antimicrobial activity; Bleaching treatment; Characterization; Jackfruit skin powder fibre; Polylactic acid

	Carofiglio, et al.	2017	Novel PHB/Olive mill wastewater residue composite based film: Thermal, mechanical and degradation properties	Bioplastic; Degradation; Polyhydroxybutirrate (PHB); Wastewater
Composite food packaging biobased plastics food waste	Mohanty, et al.	2018	Composites from renewable and sustainable resources: Challenges and innovations	
Composite food packaging biopolymers agricultural waste	Liu, et al.	2021	A review of cellulose and its derivatives in biopolymer-based for food packaging application	Cellulose packaging; Derivatives of cellulose; Food preservation; Natural polymers
	Łopusiewicz, et al.	2021	Preparation and characterization of carboxymethyl cellulose-based bioactive composite films modified with fungal melanin and carvacrol	Agricultural residues; Antimicrobial activity; Antioxidant activity; Bioactive films; Carboxymethyl cellulose; Carvacrol; Func-tional films; Melanin
	Hoffmann, et al.	2019	Potentials nanocomposites in food packaging	
Composite food packaging bioplastics agricultural waste	Coppola, et al.	2021	Bioplastic from Renewable Biomass: A Facile Solution for a Greener Environment	Biodegradable polymers; Biomaterials; Bioplastic; Biopolymer; Environmental Pollutiion
	Ncube, et al.	2020	Environmental impact of food packaging materials: A review of contemporary development from conventional plastics to polylactic acid based materials	Biodegradable; Bioplastics; Composites; Food packaging; Polylactic acid
Composite food packaging postharvest waste	Vithu, et al.	2019	Post-Harvest Processing and Utilization of Sweet Potato: A Review	cleaning; drying; grading; peeling; product development; Sorting; starch; storage; waste and by-products utilization
	Hemalatha, et al.	2017	Efficacy of chitosan films with basil essential oil: perspectives in food packaging	Antimicrobial action; Basil oil; Chitosan; Food packaging; Squid pens

3.2. Bioplastics and biobased plastics

According to European Bioplastics, bioplastics comprise a whole family of materials that are classified according to bio-based content and biodegradability in the end of life. The term “bio-based” means that the material could be partially derived from biomass like plants (e. g. cellulose, corn, sugarcane) and this content doesn’t ensure biodegradable properties that depend on chemical structure. Consequently, a bioplastic could be fully or partially biobased and biodegradable (e. g. PLA, PHA’s, PBS), fossil derived and biodegradable (e. g. PBAT), such as partially biobased but no biodegradable (e. g. bio-PET, bio-PP, etc) (Bioplastics, 2018). The two thermoplastic polyesters polyhydroxyalkanoates PHA’s, polylactic acid PLA and them composites are considered the most favoured bioplastics to replace fossil-based polymers for food packaging applications (Boey, Mohamad, Khok, Tay, & Baidurah, 2021) (Ncube, Ude, Ogunmuyiwa, Zulkifli, & Beas, 2020).

PHA’s are a family of thermoplastic polymer synthesized by bacteria fermentation and extraction procedures from different carbon substrates. Carbon sources can be derived also from organic waste as milk and sugar processing industries (Saratale, Cho, Dattatraya Saratale, & Kadam, 2021). Especially sugar molasses is characterized by high amount of glucose and fructose, the monomers contained in sucrose and needed resources for intracellular synthesis (Boey, Mohamad, Khok, Tay, & Baidurah, 2021).

Polylactide or polylactic acid PLA, is synthesized by polycondensation of lactic acid or by ring -opening polymerization (ROP) of lactide; lactic acid which can be obtained by microbial fermentation of sugarcane bagasse, whey or molasses. Due to the no toxic and biodegradable nature, (Boey, Mohamad, Khok, Tay, & Baidurah, 2021) assessed that PHA’s and PLA attracted interest in the food packaging application due to their good barrier properties toward oxygen, water vapour and the overall migration level below the migration limits for food contact materials. PHB demonstrated several drawbacks such as brittle and low toughness, while PLA is characterized by low crystallization and poor thermal stability (Boey, Mohamad, Khok, Tay, & Baidurah, 2021) (Kharrata, et al., 2020). However, PLA/PHB blend with different blending ratios, has increased the crystallinity of PLA, improved oxygen barrier and showed a comparable tensile strength to the neat PLA films.

(Qian, Zang, Wenchao, & Sheng, 2018), (Sung, Chang, & Han, 2017), (Arrieta, Peponi, Lopez, & Fernandez-Garcia, 2018) and (Herrera, Roch, & Salaberria, 2017) studied based PLA composite films reinforced with nanoparticles extracted from bamboo cellulose nano whiskers, coffee silver skin, yerba mate residue and yellow lobster shell waste. Their studies revealed that the addition of nanocrystals or nanoparticles extracted from natural fibred enhanced Young's modulus and tensile strength of composite films, produced by solution casting, compression moulding and extrusion film blowing (Jamróz, Kulawik, & Kopel, 2019). Based on these premises, (Kharrata, et al., 2020) manufactured by extrusion blowing, a PLA based thermoplastic resin compounded with date palm leaves (Phoenix dactilyfera), which residues are generated after date fruit harvesting, in semi-arid region like Tunisia. The addition of this lignocellulosic residue up to 2 wt. % with an optimum of 1% wt. promoted an increase of the apparent Young's modulus, the tensile strength and the elongation at break, of about +29 % and +12 %, respectively, along the MD direction. Above 2% wt. of natural fibers, they assessed those mechanical properties decreased because a lack of crystallinity, thermal degradation and low cohesion between matrix and fillers.

(Cinelli, et al., 2019) processed polyhydroxyalkanoate PHA with biodegradable plasticizers and variable content of natural fibers derived from food wastes as peas and wood residues; then, the composite polymers were manufactured by extrusion and injection moulding. A content up to 30% wt. allowed to enhance elastic modulus, while as the research mentioned above, tensile strength and elongation at break decreased with the amount of natural fibers due to the low adhesion between matrix and fillers. However, the obtained composite materials meet the mechanical requirements for rigid food packaging.

(Ortiz-Barajas, Arévalo-Prada, Fenollar, Rueda-Ordóñez, & Torres-Giner, 2020) melt-compounded polylactide matrix with torrefied coffee husk flour from coffee industry which was extruded and shaped by injection moulding. The content of 20% wt. of coffee fillers improved hardness, thermomechanical resistance and balanced tensile strength compared to content up to 50% wt. which decreased temperature of thermal degradation more than 7 °C, ductility, and toughness of PLA. (Songtipya, Limchu, Phuttharak, Songtipya, & Kalkornsurapranee, 2019) studied PLA/polybutyleneadipate terephthalate (PBAT) matrix added with spent coffee ground and tea leaves. The results shown that the addition of that fillers reduced overall migration values of chemicals, because food simulants didn't diffuse through composite PLA/PBAT blend added by food residues.

(Benito-González, López-Rubio, & Martínez-Sanz, 2018) assessed composite thermoplastic starch (TS) composite film, reinforced with lignocellulosic fractions extracted from Posidonia oceanic leaves. Hemicellulose and lignin were removed from the fillers composed by purified cellulose and this separation demonstrated a higher crystallinity and thermal stability for the composite polymer. The addition of pure cellulose took to optimal results in term of mechanical properties: an increase of 85% in elastic modulus and 38% in tensile strength.

Typically, the initial costs to produce bioplastics and the extraction method of fillers are high. (Ahmad Marzuki, Amin Tawakkal, Mohd Basri, & Othman, 2020) studied low-cost fibre reinforced polylactic acid with the aid of bleaching treatment and using tropical food waste as jackfruit skin, largely consumed in the Southeast Asia. The addition of only natural fibres didn't improve tensile performance yet bleaching treatment of that material, with a content up to 30% wt. increased its mechanical and thermal properties. Furthermore, the insertion on a basil content of 15% wt. demonstrated antimicrobial properties, with a lower growth of gram-positive bacteria compared to the non-treated fibres.

In the work of (Carofiglio, et al., 2017), PHB was blended with olive mill residual from production and refinery of olive oil, a low-cost resource of fine chemicals rich in lignin cellulose and polyphenols. It was demonstrated that the increasing of fillers concentration improved thermal properties of the packaging, useful in the degradation process in the end of life in aqueous and soil environment. Therefore, (Petinakis, Liu, Yu, Way, & Sangwan, 2010) compared the degradation rate in composting test between pure PLA and PLA blended with natural fibres as starch or wood flour. They founded that the addition of hydrophilic natural fibres accelerated the biodegradation rate up to 80% after 80 days, when the starch concentration is increased from 10% to 40%.

3.3 Biopolymers and biobased polymers

The current paragraph investigates the results of review analysis referred to alternative biopolymers derived from renewable resources. (Otonia, Lodi, Lorevice, Leitãoc, & Ferreiraa, 2018) investigated the production of a biodegradable bio-composite made of hydroxypropyl methylcellulose (HPMC) and micro fluidized cellulose fibers with the adding of carrot processing waste. A content of 33% wt. of carrot residue allowed to obtain mechanical properties, as Young's modulus and elongation at break in accordance with the requirement for food packaging uses.

Bioactive films were manufactured by (Łopusiewicz, et al., 2021), using carboxy methyl cellulose and addition of carvacrol and fungal melanin extracted from mushrooms derived from agro-industrial wastes. The addition of melanin enhanced UV protection, water vapour barrier, antioxidant, and mechanical properties while, carvacrol improved antimicrobial properties against Escherichia coli, Staphylococcus aureus, and Candida albicans. However, a lack of transparency occurred due to the content of carvacrol (Hoffmanna, Amaral Petersa, & Angioletti, 2019). Wastes as potato peels and sweet lime pulp derived from juice extraction, were used in several ratio in the production of composite films. The forming solution was processed with ultrasound treatment to reduce the dimension of particles, allow the blending process increasing film strength and elongation properties (Prasad Borah, Das, & Badwaik, 2017). (Hemalatha, UmaMaheswari, Senthil, Krithiga, & Anbukkarasi, 2017) in their publication studied chitosan matrix derived from squid pens compounded with essential oil as basil oil. They founded that basil oil at 0,5% concentration, allowed the fully inhibition of food pathogenic fungi as Aspergillus flavus, Fusarium sp. and Penicillium sp. and an increase of barrier properties. However, the incorporation of basil oil on this edible film a low tensile strength and elongation at break.

4. Conclusions

The present work investigated the high potential application of agricultural, food and postharvest waste as fillers in the manufacturing of composite food packaging. The research of bio-composites, even from waste residuals is taking attention on scientists and researchers that are starting to discover sustainable low cost, low greenhouse gas emissions materials with improved performance for food packaging applications. This brief literature analysis that considers very recent publication, demonstrated that thermoplastics polymers as PHAs, PLA in several blending and added with different fillers recovered by waste residue, have improved their physical, mechanical, and biodegradable properties.

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