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PRODUÇÃO E CARACTERIZAÇÃO DE FILMES ATIVOS ANTIOXIDANTES E AROMÁTICOS À BASE DE CAQUI (*Diospyros kaki* L.) E FARINHA DE CASCA DE LARANJA (*Citrus sinensis)*

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Dissertação de Mestrado apresentada ao Programa de Pós-Graduação em Alimentos e Nutrição (PPGAN) da Universidade Federal do Estado do Rio de Janeiro (UNIRIO), como parte dos requisitos necessários à obtenção do título de Mestre em Alimentos e Nutrição.

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Prof. Dr. Ricardo Felipe Alves Moreira

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“Foi o tempo que dedicaste à tua rosa que a fez
tão importante.”

Antoine de Saint-Exupéry

RESUMO

Filmes ativos antioxidantes e aromáticos de purê de caqui (*Diospyros kaki* L.) (1000 – 880 g/kg de solução filmogênica) incorporados com farinha de casca de laranja (*Citrus sinensis*) (40 – 0 g/kg de solução filmogênica) e glicerol (80 – 0 g/kg de solução filmogênica) foram desenvolvidos pela técnica de *casting* e caracterizados. Os filmes apresentaram 0,2 – 1,71 MPa, 10.24 – 15.84% e 1.8 – 15.32 MPa de resistência à tração (TS), alongamento na ruptura (EB) e módulo de Young (YM), respectivamente. Os filmes desenvolvidos obtiveram altos valores de permeabilidade ao vapor de água (7.09 – 12.61 × 10^{-6} g h^{-1} m^{-1} Pa^{-1}) e solubilidade em água (WS) (45.27 – 75.80%). Os filmes apresentaram coloração alaranjada semelhante à polpa madura do caqui. A microscopia eletrônica de varredura mostrou filmes com superfície compacta e homogênea. A análise termogravimétrica evidenciou duas etapas importantes de degradação relacionadas ao glicerol e pectina (130°C - 230°C) e de decomposição da estrutura do polímero (acima de 300°C). A espectroscopia de infravermelho por transformada de Fourier mostrou picos de absorção semelhantes entre as amostras, entretanto, interações entre componentes do glicerol e da pectina foram observadas resultando em alterações na magnitude. Os ensaios de ABTS revelaram capacidade antioxidante dos filmes que variou de 919.9 a 2747 µg/mL IC_{50}, além de um conteúdo total de fenólicos que variou de 3.8 a 5.3 mg EAG/g. Um total de 71 compostos voláteis pertencentes a diferentes classes químicas foram identificados, incluindo limoneno, β-ionona e o ácido vacênico, que influenciam no aroma geral e nas propriedades bioativas dos filmes. Os dados obtidos demonstram o potencial dos filmes desenvolvidos como material ativo para embalagens flexíveis e coloridas de alimentos. Entre as formulações produzidas, aquela com 98% de purê de caqui e 2% de farinha de casca de laranja foi a melhor, porque apresentou propriedades mecânicas que se destacaram entre as demais: 1.71 MPa, 10.5% e 15.32 MPa de TS, EB e YM, respectivamente, além de uma menor solubilidade em água (55.23%) e um melhor potencial antioxidante comparado aos demais filmes desenvolvidos (919.9 µg/mL IC_{50}). Seu perfil aromático apresentou 60 compostos voláteis diferentes, sendo o limoneno o composto terpênico mais expressivo [(1.2)%], possuindo limiar de odor em água de 10 µg L^{-1}. Este estudo traz uma proposta que contribui para reduzir as perdas pós-colheita de caqui e reaproveitar os subprodutos cítricos afim valorizá-los ao gerar produtos de valor agregado, estando em linha com os preceitos da economia circular.

Palavras-chave: Aproveitamento de resíduos; Filmes compósitos; Propriedade de barreira e mecânica; Compostos bioativos; Embalagem de alimentos; Economia Circular.

ABSTRACT

Antioxidant and aromatic active films of persimmon puree (*Diospyros kaki* L.) (1000 – 880 g/kg of film-forming solution) incorporated with orange peel flour (*Citrus sinensis*) (40 – 0 g/kg of film-forming solution), and glycerol (80 – 0 g/kg of film-forming solution) were developed by the casting technique and characterized. The films presented 0.2 – 1.71 MPa, 10.24 – 15.84%, and 1.8 – 15.32 MPa of tensile strength (TS), elongation at break (EB), and Young's modulus (YM), respectively. The developed films had high values of water vapor permeability (7.09 – 12.61 × 10^{-6} g h^{-1} m^{-1} Pa^{-1}) and solubility in water (WS) (45.27 – 75.80%). The films showed an orange color similar to the mature pulp of persimmon. Scanning electron microscopy showed films with a compact and homogeneous surface. Thermogravimetric analysis showed two important degradation steps related to glycerol and pectin (130°C - 230°C) and polymer structure decomposition (above 300°C). Fourier transform infrared spectroscopy showed similar absorption peaks between samples; however, interactions between glycerol and pectin components were observed, resulting in changes in magnitude. The ABTS tests revealed the antioxidant capacity of the films that ranged from 919.9 to 2747 µg/mL IC_{50}, in addition to a total phenolic content that ranged from 3.8 to 5.3 mg EAG/g. 71 volatile compounds belonging to different chemical classes were identified, including limonene, β-ionone, and vacenic acid, which influence the films' overall aroma and bioactive properties. The data obtained demonstrate the films' potential as an active material for flexible and colored food packaging. Among the formulations produced, the one with 98% persimmon puree and 2% orange peel flour was the best because it presented mechanical properties that stood out among the others: 1.71 MPa, 10.5%, and 15.32 MPa of TS, EB, and YM, respectively, in addition to a lower solubility in water (55.23%) and a better antioxidant potential compared to the other films developed (919.9 µg/mL IC_{50}). Its aromatic profile showed 60 different volatile compounds, with limonene being the most expressive terpene compound [(1.2)%], with an odor threshold in the water of 10 µg L^{-1}. This study brings a proposal that contributes to reduce post-harvest losses of persimmon and reuse citrus by-products in order to enhance them by generating value-added products, in line with the precepts of the circular economy.

Keywords: Waste Utilization; Composite Films; Barrier and Mechanical Properties; Bioactive Compounds; Food Packaging; Circular Economy.

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SUMÁRIO

1. INTRODUÇÃO

À medida que aumenta a consciência sobre a importância de promover impactos positivos que colaboram para reconstituir o meio ambiente e melhorar a qualidade de vida de forma geral, a indústria se vê forçada a repensar o modelo tradicional de "descarte" dos materiais gerados comumente relacionados à economia linear (SHOGREN et al., 2019). Em contraponto, um novo modelo econômico e de desenvolvimento sustentável ganha visibilidade: a economia circular. A Economia Circular é um conceito que repensa as práticas econômicas e objetiva manter produtos, componentes e materiais em circulação tirando proveito do máximo de valor e utilidade entre ciclos técnicos e biológicos, inspirada nos conceitos cíclicos da própria natureza. A indústria de embalagens desempenha um papel crucial nessa mudança de paradigma uma vez que movimenta bilhões de dólares todos os anos (MEHERISHI; NARAYANA; RANJANI, 2019) e continua a depender majoritariamente de materiais ambientalmente pouco sustentáveis como plásticos de origem fóssil de uso único e embalagens multicamadas, que representam cerca de 15-20% de resíduos sólidos em diferentes países (HALL, 2017; TENCATI et al., 2016).

Cabe mencionar que diante da maior preocupação com a higiene, da necessidade de uso de EPI's para conter a transmissão de Sars-CoV-2 e com o crescimento expressivo de compras online durante a pandemia de COVID-19 percebeu-se um aumento no consumo de materiais e de embalagens plásticas *single use* e um consequente retrocesso de agendas globais como o "Compromisso Global para a Nova Economia dos Plásticos". Muitas das políticas destinadas a limitar produtos plásticos de uso único foram revertidas e o desenvolvimento de embalagens de fontes renováveis, biodegradáveis e compostáveis é urgente, como parte das ações para conter a poluição plástica (BARONE et al., 2021; ELLEN MACARTHUR FOUNDATION, 2021).

Imbuída no contexto de embalagens que circulem de forma positiva nos nossos sistemas, - design circular do berço ao berço – as tendências de mercado apontam para o desenvolvimento de embalagens biodegradáveis à partir de excedentes vegetais e resíduos agroindustriais (MEYS et al., 2020; LUTTENBERGER, 2019). Nesse sentido, sabe-se que a América Latina apresenta um dos maiores índices de perdas de frutas e hortaliças, onde o maior percentual está relacionado com as etapas de pós-colheita e processamento (FAO, 2015). Inserido nesse cenário, destaca-se o caqui (*Diospyros kaki* L.), fruto com grande volume de produção no Brasil e que apresenta uma composição química rica em carboidratos, fibras e compostos ativos com propriedades antioxidantes como os polifenois, carotenoides e taninos

(GEA-BOTELLA et al., 2021; MATHEUS et al., 2020), evidenciando alternativas promissoras para sua utilização e valorização em diversos segmentos, inclusive para o desenvolvimento de bioplásticos (MATHEUS et al., 2021; RAMACHANDRAIAH; GNOC; CHIN, 2017). Cabe ainda ressaltar que embora o Brasil seja o maior produtor de caqui de toda América, ocupando a quinta posição mundial, observa-se uma expressiva perda pós-colheira relacionada a esta fruta (FAOSTAT, 2019), reforçando a necessidade de se estudar novas rotas de aproveitamento que visem agregar valor à este produto.

Os resíduos cítricos, por sua vez, são globalmente abundantes e desafiadores já que são materiais quimicamente interessantes e subutilizados (BÁTORI et al., 2017). Entre as frutas cítricas, as laranjas são as frutas tropicais mais comumente produzidas em todo mundo (FARAHMANDFAR et al., 2020), alcançando uma taxa de produção anual de cerca de 78 milhões de toneladas em 2019 (FAOSTAT, 2020). Durante o processamento industrial para produção de sucos, a laranja gera em média 50% de resíduo em relação à sua massa total (em Kg), correspondente à massa da casca (BÁTORI et al., 2017). Esse resíduo contêm alto teor de pectina (20 – 30%), fibras dietéticas, celulose, hemicelulose, amido, lignina, flavonoides e óleos essenciais (FARAHMANDFAR et al., 2020; VENKATESH; SUTARIYA, 2019) e vêm se mostrando interessante em diversas aplicações tecnológicas, inclusive como aditivo de reforço mecânico e de veículo de bioativos em plásticos de base biológica (LUCHESE; PAVONI; TESSARO, 2021; KEVIJ et al., 2020).

Esta dissertação está estruturada em três capítulos, escritos sob forma de artigo científico, que versam sobre a produção de filmes ativos com um olhar atento para o contexto da COVID-19. O primeiro capítulo intitulado: "***Green-based active packaging: Opportunities beyond COVID-19, food applications and perspectives in circular economy – a brief review***", é uma revisão bibliográfica sobre as tendências e questões econômicas relacionadas a materiais biodegradáveis para embalagens de alimentos; o desenvolvimento e aplicação de filmes ativos de base biológica; algumas oportunidades além do COVID-19 para o segmento de embalagens de alimentos e perspectivas na economia circular. **Este artigo foi aceito e publicado pela revista Comprehensive Reviews in Food Science and Food Safety (ISSN: 1541-4337) (DOI: 10.1111/1541-4337.12812).**

O segundo capítulo intitulado: "***Development of biodegradable food packaging in the context of COVID-19: sustainability more urgent than ever***" consiste em uma breve revisão bibliográfica abordando as principais conquistas, desafios e perspectivas das embalagens de alimentos biodegradáveis de base biológica e tendências futuras no contexto do COVID-19.

Este artigo está submetido na revista Sustainability Management Forum (ISSN: 2522-5987).

Por fim, o terceiro e último capítulo intitulado: "***Active antioxidant and aromatic films based on persimmon (Diospyros kaki L.) and orange peel flour (Citrus sinensis)***" é um artigo sobre o desenvolvimento e caracterização de filmes ativos antioxidantes e aromáticos de purê de caqui incorporados de farinha de casca de laranja e glicerol. Os filmes foram caracterizados quanto suas propriedades mecânicas, físicas, físico-químicas, ópticas, antioxidantes, conteúdo de fenólicos totais e perfil de compostos voláteis. **Este artigo será submetido para a revista Current Research in Food Science (ISSN: 2665-9271).**

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2. OBJETIVOS

2.1 GERAL

Desenvolver filmes ativos antioxidantes e aromáticos à base de caqui (*Diospyros kak*i L.) e farinha de casca de laranja (*Citrus sinensis*).

2.2 ESPECÍFICOS

- Desenvolver uma revisão descritiva sobre o desenvolvimento e aplicação de filmes ativos e biodegradáveis para embalagem de alimentos no contexto da COVID-19 e da economia circular;
- Desenvolver uma revisão descritiva sobre conquistas, desafios e perspectivas futuras das embalagens de alimentos biodegradáveis a partir da COVID-19;
- Desenvolver diferentes formulações de filmes à base de purê de caqui (*Diospyros kaki* L.) incorporados de farinha de casca de laranja (*Citrus sinensis*) e glicerol;
- Caracterizar os filmes desenvolvidos quanto suas propriedades de barreira, mecânicas, físico-químicas e ópticas;
- Avaliar o potencial antioxidante dos filmes;
- Avaliar o conteúdo de compostos fenólicos dos filmes;
- Identificar e quantificar o perfil de compostos voláteis dos filmes.

CAPÍTULO I

Este artigo foi aceito e publicado pela revista Comprehensive Reviews in Food Science and Food Safety (ISSN: 1541-4337) (DOI: 10.1111/1541-4337.12812).

Green-based active packaging: opportunities beyond COVID-19, food applications and perspectives in circular economy - a brief review

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Title Short Version: Green-based active packaging

ABSTRACT

The development of biodegradable packaging, based on agro-industrial plant products and by-products, can transform waste into products with high added value and reduce the use of conventional non-renewable packaging. Green-based active packaging has a variety of compounds such as antimicrobials, antioxidants, aromatics, among others. These compounds interact with packaged products to improve food quality and safety and favor the migration of bioactive compounds from the polymeric matrix to food. The interest in the potential hygienic-sanitary benefit of these packages has been intensified during the COVID-19 pandemic, which made the population more aware of the relevant role of packaging for protection and conservation of food. It is estimated that the pandemic scenario expanded food packaging market due to shift in eating habits and an increase in online purchases. The triad health, sustainability, and circular economy is a trend in the development of packaging. It is necessary to minimize the consumption of natural resources, reduce the use of energy, avoid the generation of waste and emphasize the creation of social and environmental values. These ideas underpin the transition from the emphasis on the more subjective discourse to the emphasis on the more practical method of thinking about the logic of production and use of sustainable packaging. Presently, we briefly review some trends and economic issues related to biodegradable materials for food packaging; the development and application of bio-based active films; some opportunities beyond COVID-19 for food packaging segment, and perspectives in circular economy.

Keywords: Active packaging; Sustainable; Food security; Bioplastic; Agroindustrial waste.

1. Introduction

Conventional plastics are among the materials currently used in food packaging today and play an important role due to their durability, resistance and performance/cost ratio (Geyer et al. 2017). However, these properties represent a severe threat to the environment as plastics do not naturally degrade to a large degree (Shen et al. 2020). In addition, most plastics have a very short life cycle, i.e., they are used only once and for a few minutes (Geyer et al. 2017). This has become a significant challenge on the waste management process, mainly due to the increase in white pollution, which refers to an image denomination for the phenomenon of environmental pollution from plastic waste (Dauvergne, 2018). It is estimated that around 280 million tons of plastic waste were produced in 192 countries and coastal regions in 2011, and approximately 8 million tons of waste flowed into the oceans (Shen et al. 2020), representing a significant disadvantage in the post-use phase of packaging (Petlzer et al. 2017).

To reduce this problem, technological advances allowed the development of biodegradable films using materials from renewable and non-polluting sources, and agribusiness residues are the first-choice sources of raw material for producing these films (Matheus et al. 2021). The use of different types of food waste as a natural source of polymers, produces films that can be reincorporated into the environment within a short period of time, and it also reduces waste accumulation, leading to positive impacts on the environment (Zhong et al. 2020).

Biodegradable films can also be considered as a type of active packaging which, by definition, intentionally interacts with the packaged food and promotes improvements, especially regarding the sensory and nutritional quality of the packaged products (Bhardwaj et al. 2019). In practice, active packaging refers to the incorporation and/or the immobilization of certain bioactive compounds in the polymeric matrix, allowing it to interact with the food and the environment, offering innovative solutions to maintain or prolong the useful life, improve, and monitor the quality and safety of food (Almasi et al. 2020). The main properties exhibited by active packaging are related to substances that absorb oxygen, ethylene, moisture, and odor, and those that contain antimicrobial, antioxidant and aromatic agents (Wyrwa & Barska, 2017). The importance of packaging for food safety concerning hygienic-sanitary aspects has gained greater attention over the years. The COVID-19 pandemic has reinforced, especially to the population, the great functionality of packaging as a fundamental protection to food. There was a growing trend in the use of plastic packaging for food (Vanapalli et al. 2021), associated with a greater purchase and storage of processed food products (Grand View Research, 2020a), and increased delivery of meals that depend on disposable packaging (Southey, 2020; Vanapalli et al. 2021). In this sense, it becomes relevant to invest in the development of biodegradable and active packaging that is more competitive with the conventional packaging market. This effort contributes to decreasing the amount of setbacks suffered by environmental sustainability, to the detriment of health-oriented initiatives.

Thus, the objective of the present study was to carry out a bibliographic review on active green-based packaging, to explore opportunities beyond the COVID-19 pandemic, applications in food and perspectives in the circular economy.

2. Methodology

This bibliographic review was carried out using a qualitative approach. The data were obtained through research in scientific articles indexed in the Pubmed, Web of Science, Google Scholar, and Scopus electronic databases. The inclusion criteria comprised: full articles available, which were consistent with the theme of the study on screen and exclusively related to the topics covered.

3. Scientific production related to active packaging and circular economy – a rise in the 21st century

The interest in the study of active packaging and circular economy has grown over the past few years. Figure 1 shows the number of scientific articles published between the years 2000 to 2020. Considering the Scopus database, the main countries that have published articles about active packaging in the past years are the United States (1046 articles), China (555 articles), Spain (504 articles), Italy (394 articles), and Brazil (325 articles); and the main journals are Food Packaging and Shelf Life (167 articles), Food Hydrocolloids (140 articles), International Journal of Biological Macromolecules (136 articles), Packaging Technology and Science (97 articles), and Carbohydrate Polymers (96 articles). Regarding the circular economy, the countries with the highest number of publications in the past years are China (1428 articles), Italy (1162 articles), United Kingdom (992 articles), Spain (777 articles), and United States (629 artciles); and the main journals are Journal of Cleaner Production (658 artciles), Sustainability Switzerland (494 articles), Resources Conservation and Recycling (279 articles), and Waste Management (149 articles).

Figure 1. Number of publications of scientific articles about active packaging (a) and circular economy (b) from 2000 to 2020.

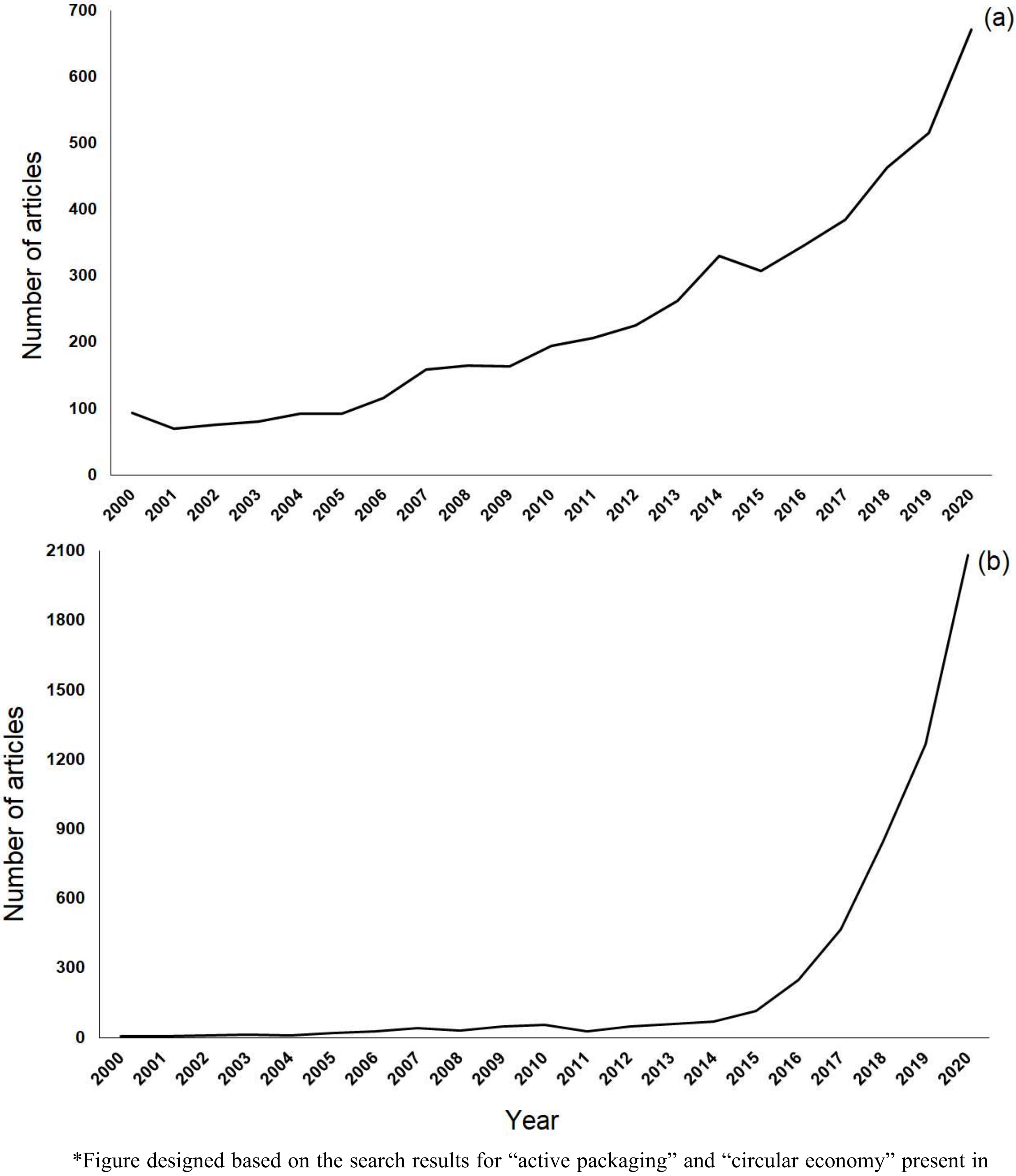


*Figure designed based on the search results for "active packaging" and "circular economy" present in the Title, Abstract, and/or Keywords of the articles, in the Scopus database.

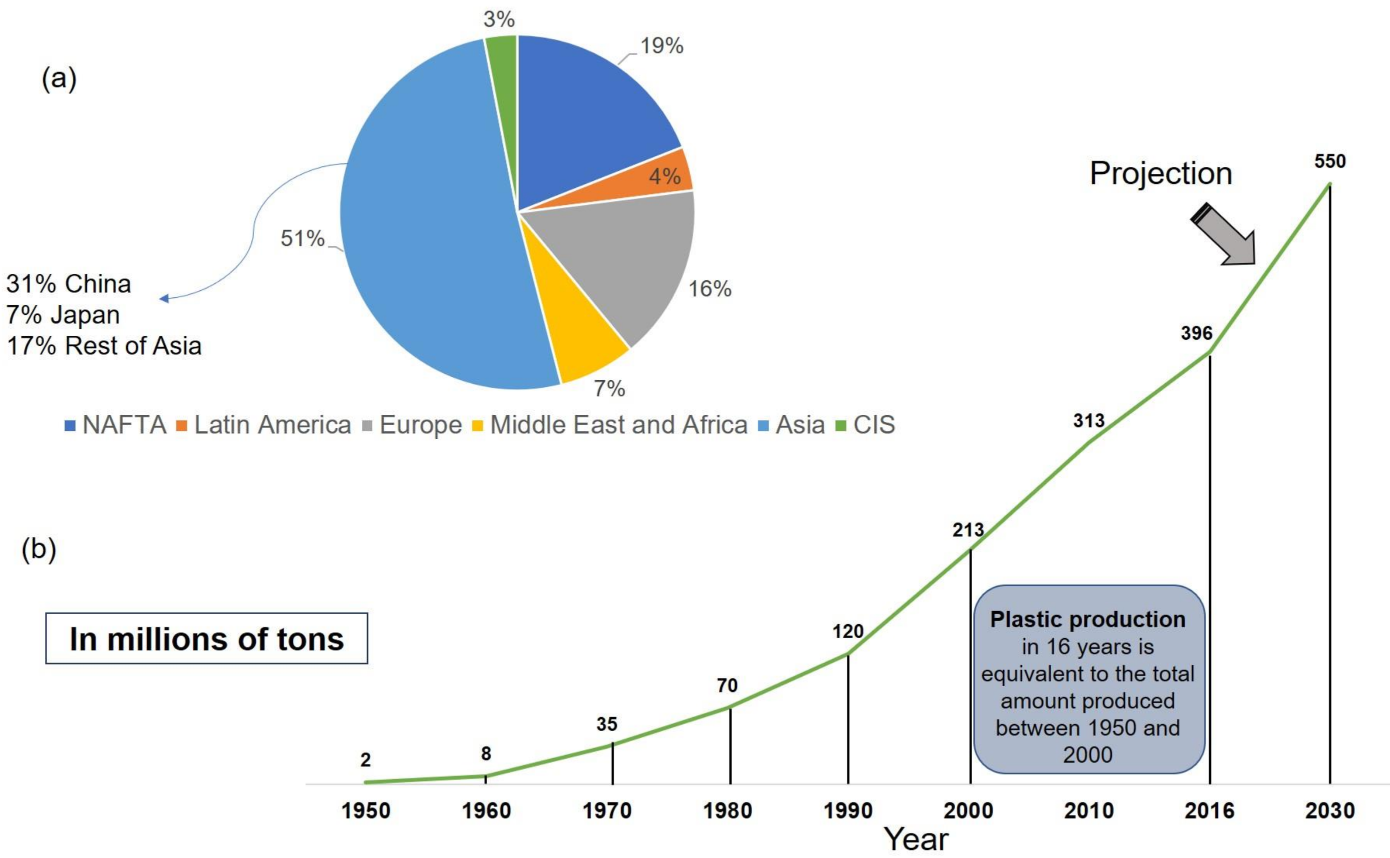
The recent increase in interest in these issues has been stimulated globally by the need to redesign products, processes, and services that use raw materials sustainably and seek to reduce waste (Foschi & Bonoli, 2019). In addition, the COVID-19 pandemic has led to an increase in the need for food quality and safety, and efforts have been made to understand and improve the active role of food packaging within this context (Vanapalli et al. 2021). In this sense, working towards common goals, this agenda is gaining visibility in governmental spheres, such as in the European Union, through the Circular Economy Action Plan, which aims to achieve ambitious targets for plastic waste management by 2030 (Foschi & Bonoli, 2019).

4. Renewable materials for food packaging: overview, economic issues and opportunities beyond COVID-19

Packaging is a key element in the trade of goods, and it can help ensure the quality of various food products (Wyrwa & Barska, 2017). The main function of packaging is to serve as a container for food, allowing satisfactory transport throughout the supply chain, and acting as a barrier to protect food from the influence of environmental factors, such as oxygen, moisture, light, chemical and microbiological contamination (Yildirim et al. 2018). The growing race to produce knowledge about the importance of packaging, contributes to the constant improvement of production methods in the industry (Wyrwa & Barska, 2017).

The global plastic production market has been growing at a fast pace decade after decade, with astronomical projections for the future, as shown in Figure 2.

Figure 2. Economic data on the global plastics sector. (a): Distribution of global plastics production by continent or economic bloc (NAFTA: The North American Free Trade Agreement and CIS: The Commonwealth of Independent States). (b): Accelerated growth of plastic production in millions of tons and projection for 2030.



Sources: Plastics Europe Associaton of Plastics Manufacturers (2020); Revista pesquisa Fapesp (2019).

According to Plastics Europe (2020), the main application of plastics is as packaging material, accounting for 39.6% of total European production, which reached 50.7 million tons in 2019. Within this large sector, food packaging has received a great deal of attention with a global market estimated at around US $ 303 billion in 2019 and a 5.2% growth forecast until 2027, with an increase, above all, for flexible packaging. Thanks to the characteristics of this material in forming thin, light and compact packaging, the growth of this sector is expected to be more pronounced when compared to rigid packaging, for example (Grand View Research, 2020a).

In general, plastic packaging has characteristics such as low production cost, good mechanical performance, lightness, and a satisfactory oxygen barrier (Carwford & Martin, 2020). However, most of these polymers are of petrochemical origin, and the increase in their consumption inevitably results in socioeconomic problems, e.g., scarcity and rising oil prices, and environmental problems, such as the generation and accumulation of solid waste, which can take hundreds of years to decompose (Geyer et al. 2017).

In addition to the trend towards increased use of plastics because of their characteristics, an important issue that must be addressed is the broader application of plastics in packaging as a result of the lockdown measures imposed by COVID-19. The increasing demand for packaged food, bottles for milk and fruit juices, crates, and caps for food packaging in the pandemic, are among the products that will push the demand for polyethylene in the next years (Grand View Research, 2020b). The global plastic packaging market size in 2019 was USD 234.14 billion, showing a compound annual growth rate (CAGR) of 4.0% from 2020 to 2027. The segments of food and beverages presented a revenue share of over 51.1% in 2019 and growth is expected during the next years (Grand View Research, 2020b).

The pandemic lockdown boosted online shopping and takeaway services, including food and groceries (Parashar & Hait, 2021). To reduce the spread of COVID-19, the distribution of those goods to the residences has raised, leading to higher usage of plastic packaging materials (Deka et al. 2020). As a result of safety concerns when going out (e.g., shopping in supermarkets, eating in a restaurant, etc.), and the preference for e-commerce purchases and takeaway services for home delivery, there has been a change in consumer preference, leading to the growth of the single-use food packaging market (Parashar & Hait, 2021; Silva et al. 2021). Owing to the raised demand for online food and groceries during the pandemic, plastic packaging waste such as polypropylene (PP), high-density polyethylene (HDPE), low-density polyethylene (LDPE), polyethylene terephthalate (PET), and polystyrene (PS) are expected to increase (Sharma et al. 2020a). According to the Global Market Insights (2020) report, the global packaging market size during 2019 was valued at USD 909.2 billion and is expected to reach USD 1,012.6 billion by 2021, recording a CAGR of 5.5% during the pandemic. Plastic is the material projected to lead the packaging market in this period.

The COVID-19 pandemic has accented the fragility of the waste management system and led to an increased dependence on single-use plastics, leading to policies aimed at plastic waste reduction. Thus, the pandemic has highlighted an old problem of overconsumption of plastics, and the need for sustainable, long-term solutions is more urgent than ever (Silva et al. 2020). To overcome resource depletion and plastic pollution, producers must invest in sustainable bioplastics produced from bio-based and biodegradable polymers; and there must be a transition from the current economy - linear economy - to a circular economy. In order to design a system that can combat future pandemics, policies that encourage the use of ecological bioplastics and circular technologies should be formulated and implemented effectively. The implementation of a circular economy model would help retain more resources in the production and consumption circle, thus reducing the amount of waste generated (Sharma et al. 2020b). This transition involves an increase in the responsibility of industries and governments as well as research and development to review the material management system considering the entire life cycle of such materials (Silva et al. 2020).

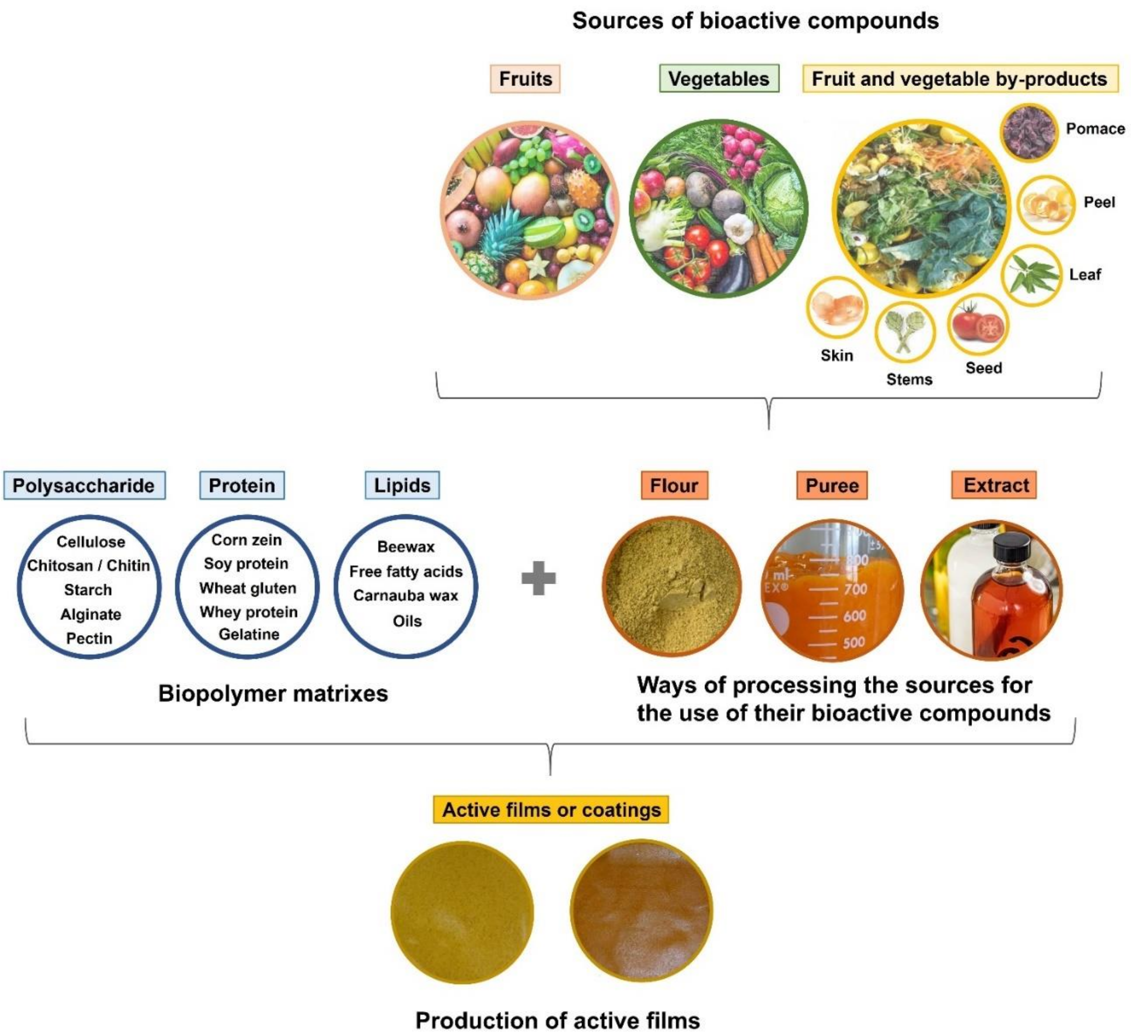
Biobased materials have been developed in order to minimize the environmental issues caused by conventional plastics. The increased awareness concerning the negative impacts of traditional plastics has been the main driver of the bioplastic packaging market (Galanakis et al. 2021). Global production of bioplastics in 2020 was estimated at 2.11 million tonnes, with 47% (0.99 million tonnes) of this volume used by the packaging industry, the largest market segment in the bioplastics industry. Global production of bioplastics is expected to reach 2.87 million tonnes by 2025. The biobased market has emerged as a sustainable solution, and even considering the concerns raised by the COVID-19 pandemic, global market growth is expected to reach 36% over the next five years (European Plastics – Bioplastics market data).

The food packaging market during and post-pandemic should provide consumers with information on sustainable practices, such as how to recycle packaging; design packaging with the hygienic-sanitary concept in mind, such as antimicrobials and antivirals, through innovative packaging (active and intelligent); design e-commerce packaging, with attractive designs and ready-to-ship templates, promoting and enhancing the consumer experience; and design packaging with the basic principles of cost, performance, and convenience in mind (Feber et al. 2020).

5. Agro-industrial wastes as renewable sources: food applications and life cycle assessment (LCA)

There is an increasing need to find new ways to use agro-industrial waste in order to reduce its environmental impact, as well as to implement new strategies for the management of agricultural waste in a circular bio-based economy. When used efficiently, agro-industrial by-products, residues and food residues broaden the spectrum of benefits, which go beyond their use as feed or for the production of biogas (Monari et al. 2020). The development of biodegradable films from by-products is an excellent strategy to add value to these residues, which are discarded in large amounts by the food industry; in addition, they are a source of bioactive compounds, e.g., polysaccharides, fibers, polyphenols, vitamins, and minerals (Dilucia et al. 2020).Figure 3 shows the application of fruits, vegetables, and their by-products to develop an active packaging.

Figure 3. Application of fruits, vegetables and their by-products to produce biodegradable active packaging.



Sources: Dilucia et al. (2020); Popović et al. (2018).

Biodegradable films, are independent structures made from flexible biopolymers that, when used directly on food, form a protective barrier between them and the surrounding environment, and can be consumed as part of the food (Otoni et al. 2017). In addition, they can be reincorporated into the environment in a short time and can be applied as packaging material in various segments of the productive sector and consumer products (Zhong et al. 2020). These polymers can be extracted directly from biomass (polysaccharides, lipids, or proteins) (Nilsen-nygaard & Sivertsvik, 2020), can be produced by chemical synthesis using monomers from renewable sources, by microorganisms or through biotechnology (Lalanne et al. 2021). However, the interest in adding value to waste from agricultural commodities arouses interest in new applications for it, especially for waste from the juice industry (Luchese et al. 2021). More than 35 species of fruits, vegetables and their by-products have been identified in the literature; they are used as raw material for formulating biodegradable packaging. This means that this field has great potential to be explored (Otoni et al. 2017). Bio-based films made from fruits and vegetables usually preserve the main characteristics of flavor, color, and aroma inherent in vegetables, and they offer a competitive advantage in terms of sensorial and nutritional qualities (Brito et al. 2019).

The integrity of our ecosystems is threatened by the increasing amount of plastic waste generated each year, owing to the growth in the consumption of disposable plastics in food packaging (Kakadellis & Harris, 2020). The use of some bio-based plastics, given their biodegradable properties, has been recognized as a sound strategy to divert food and food packaging waste away from landfills while avoiding plastic leaking into the environment. Therefore, reverting food waste into bio-based films, or using it to produce bioactive compounds which can be incorporated into food packaging, would minimize the food waste generated and result in 'greener' end-of-life scenarios such as anaerobic digestion and composting, thus contributing to a circular economy (Kakadellis & Harris, 2020).

To reduce the environmental effects of packaging systems, the use of new materials or the application of eco-design techniques have been applied. The development of these materials could be employed as short-term or single-use by replacing non-biodegradable materials extracted from fossil fuels. When designing bio-based films, environmental factors, to ensure the production of sustainable materials, must be considered. A Life Cycle Assessment (LCA) is also an essential tool to measure the environmental impact of the extraction of raw materials to ultimate disposal.

However, the LCA of biofilms is not broadly discussed in the literature (Leceta et al. 2014).

LCA is a method that aims to assess the environmental impacts of a product during its life cycle. A product has several stages of life, and each of these stages has a significant effect on the ecosphere. The LCA covers stages that start from the collection of raw material followed by processing, product manufacturing, packaging, transportation, use, maintenance, disposal and recycling (Ghosh & Katiyar, 2020). The LCA includes gathering the input and output information of a process (life cycle inventory), such as emissions, waste, and resources, and translating this information into environmental impacts, such as the contribution to climate change, eutrophication, acidification, and human and ecosystem toxicity, using standardised impact assessment methodologies (ISO 14040:2006) (Yates & Barlow, 2013; Kakadellis and Harris, 2020). By measuring the upstream materials and the downstream impacts of the product, the impact of material and process choices can be assessed, thus making it possible to evaluate the impact that the use, re-use, and ultimate disposal of a product has on the environment (Walker & Rothman, 2020). The LCA analysis calculator comprises 3 steps: (I) compilation of an inventory containing energy and material inputs and pollutants released to the environment; (II) quantification of the potential environmental effects associated with the inputs and releases identified; (III) and an analysis of the results to support decision-makers (Gontard et al. 2018).

In addition to the economic factors, the environmental aspects must also be considered when a new product is being designed. Leceta et al. (2014) produced films derived from biomass (soy protein, chitosan and agar), with the aim of adding value to agro-waste; the authors used the LCA method to assess the environmental behavior of the films. The results showed that the manufacturing stage most contributes to the environmental impact of chitosan and agar films given the higher energy consumption at this stage, while raw material extraction is the step that causes the greatest amount of contamination in soy protein films due to the high environmental cost related to land supply and soy cultivation. Ghosh & Katiyar (2020) analysed the LCA of chitosan-based biodegradable films compared to the LCA of fossil-based films and concluded that the latter has a greater environmental impact in terms of extraction of raw materials, production of carcinogens and the end-of-life cycle than biodegradable materials. Salwa et al. (2020) investigated the LCA of take-out food containers made from sago starch biocomposites reinforced with sugar palm fiber. The authors demonstrated that in relation to harmful effects, 49.3% had an impact on the resource category, 47.4% on human health and 3.38% on the ecosystem. The detrimental impact related to electricity is due to the high levels of energy consumption in the pre-treatment of raw materials and in the manufacture of packaging.

However, bio-based materials do not always have a lower environmental impact than traditional materials. Günkaya & Banar (2016) produced an environmental comparison between biocomposite films based on orange peel-derived pectin jelly-corn starch and low density polyethylene (LDPE) film. The biocomposite film showed slightly greater environmental damage than the LDPE films in the impact categories analyzed, with the exception of acidification. The most relevant harmful effects are related to the energy consumed to mold the film, and produce the modified starch and pectin gel. On the other hand, the biodegradation rate of the biocomposite film is faster (78.4%) than that of the LDPE film (40.4%), suggesting that this type of material can potentially be exploited as ecological food packaging. Leceta et al. (2013) compared the LCA of chitosan-based films to polypropylene (PP) films. The results showed that PP films exhibited a greater environmental impact compared to chitosan films in the categories of carcinogens and fossil fuels, with the extraction stage of the raw material being the most polluting; meanwhile, chitosan-based films had a greater environmental burden in the categories of respiratory inorganics, land use and minerals, with the greatest impact seen at the manufaturing stage of the film, due to the fact that it was not optimized from an industrial point of view. It is believed that the optimization of this process would help to reduce the consumption of energy and additives involved in the manufacture of these films. The composting scenario of bio-based films also has a highly positive effect on the environment compared to the end-of-life scenario of fossil-based films, thus reducing environmental pollution generated by the food packaging industry during disposal.

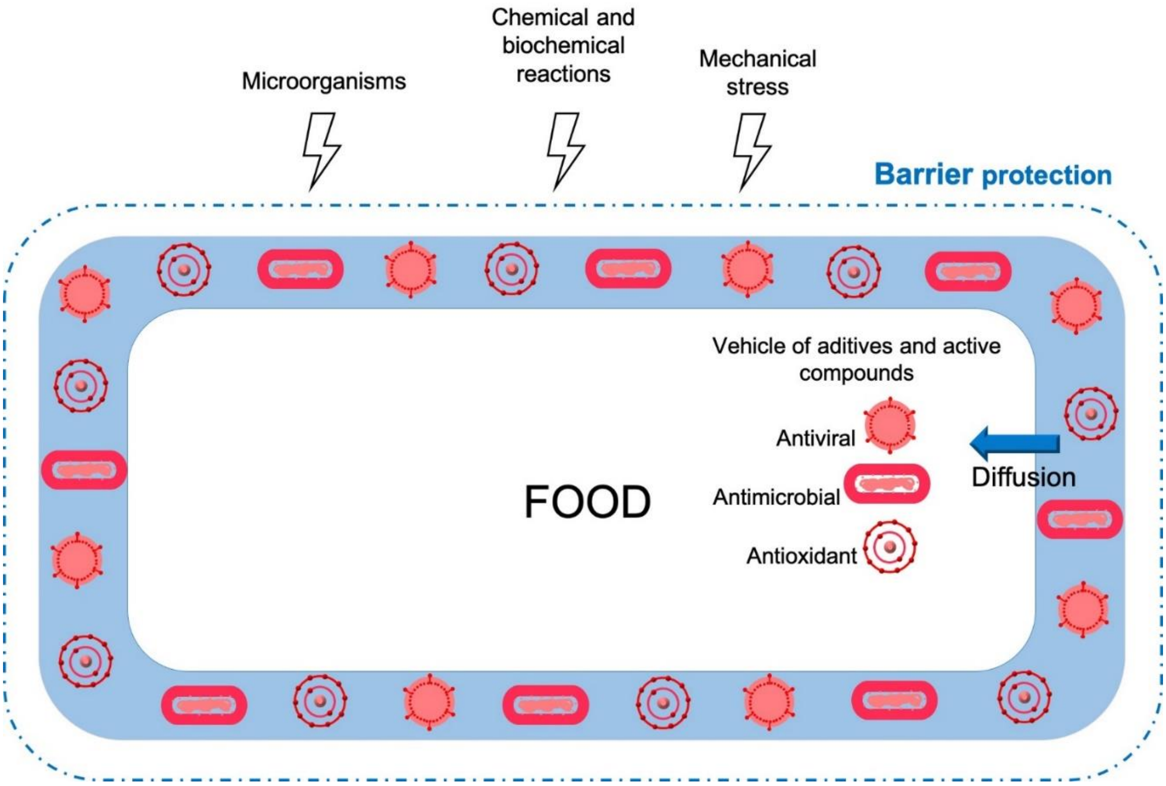
Life cycle assessment analysis is still an under-explored topic in the area of bio-based packaging from agro-residues, with most of the research focused on the use of these residues in biorefineries and in the generation of energy, such as biofuels (Ahamed et al. 2016; Lucarini et al. 2018). When exploring the value of food waste, it is fundamental to carry out a LCA of all product streams in addition to determining how to correctly employ and discard them. The exploration of agricultural by-products represents an intelligent conversion of waste into value-added products, which can be achieved through the use of green and eco-compatible technologies, in order to ensure sustainability in the supply chain. In addition, it is estimated that the transition to a green economy could generate 15 to 60 million jobs globally in the next twenty years. In this context, research that seeks to optimize the integrated recovery of these wastes is required, as well as to guide producers and consumers on virtuous and sustainable development paths (Lucarini et al. 2018; Xiong et al. 2019).

6. Development and application of active films for food packaging

Owing to its expansion in the creation of new products, the food industry has developed completely new solutions in the packaging market (Wyrwa & Barska, 2017), poses a constant challenge of meeting cosomer demands, producing modern, practical packaging that preserves food and is environmentally and economically viable (Boz et al. 2020). Before the approach of active packaging, packaging played only a passive role in the protection and commercialization of a food product; traditionally, its basic functions are divided into four categories: protection, communication, convenience, and containment (Müller & Schmid, 2019). However, new concepts of active packaging aim to play an increasingly important role, offering innovative solutions to maintain or extend the shelf life of food and improve its quality and safety (Ludwicka et al. 2020).
The concept of active packaging was originally described by Labuza and Breene (1989), as packages that intentionally interact with food, aiming to improve some of its characteristics (Wicochea-Rodríguez et al. 2019). Active packaging can be classified into two main types: (I) non-migratory packaging, for which there is no intentional migration of compounds and (II) controlled-release active packaging, which enables a controlled migration of non-volatile agents or the release of volatile compounds into the atmosphere around the food (Contreras et al. 2018). Non-migratory active packaging is based on iron oxidation, ascorbic acid oxidation, catechol oxidation or enzymatic catalysis, which work as moisture absorbers using a zeolite, cellulose and its derivatives, among others (Contreras et al. 2018; Sanches-Silva et al. 2014). This type of packaging has been applied as self-adhesive labels, sachets or incorporated into the packaging's monolayer or multilayer material (Sanches-Silva et al. 2014). Controlled-release active packaging refers to the incorporation and/or the immobilization of certain additives in

the packaging film instead of their direct incorporation into the product, allowing it to interact with both the food and the environment, playing a dynamic role in the preservation of the food product (Almasi et al. 2020). Controlled release may depend on physical and chemical phenomena, for example, the degree of affinity between the active compound and the packaging material matrix, as well as its morphology and porosity, which, at a low level, may allow a slower release (Chen et al. 2018). Bioactive agents can be incorporated into packaging films using a variety of technologies, including those in which the active agent is intended to migrate to the packaged product (Nogueira et al. 2020). The release of active compounds within food is driven by mass transfer through diffusion. This mechanism occurs when the bioactive compound diffuses across the micro or macroporous structure of the polymer, transported from the surfaces of the film to the packaged food or headspace for protective action to take place (e.g., antioxidants, antimicrobials, antivirals – see Figure 4) (Tapia-Blácido et al. 2018).

Figure 4. Release of active compounds from biodegradable films to food through diffusion.

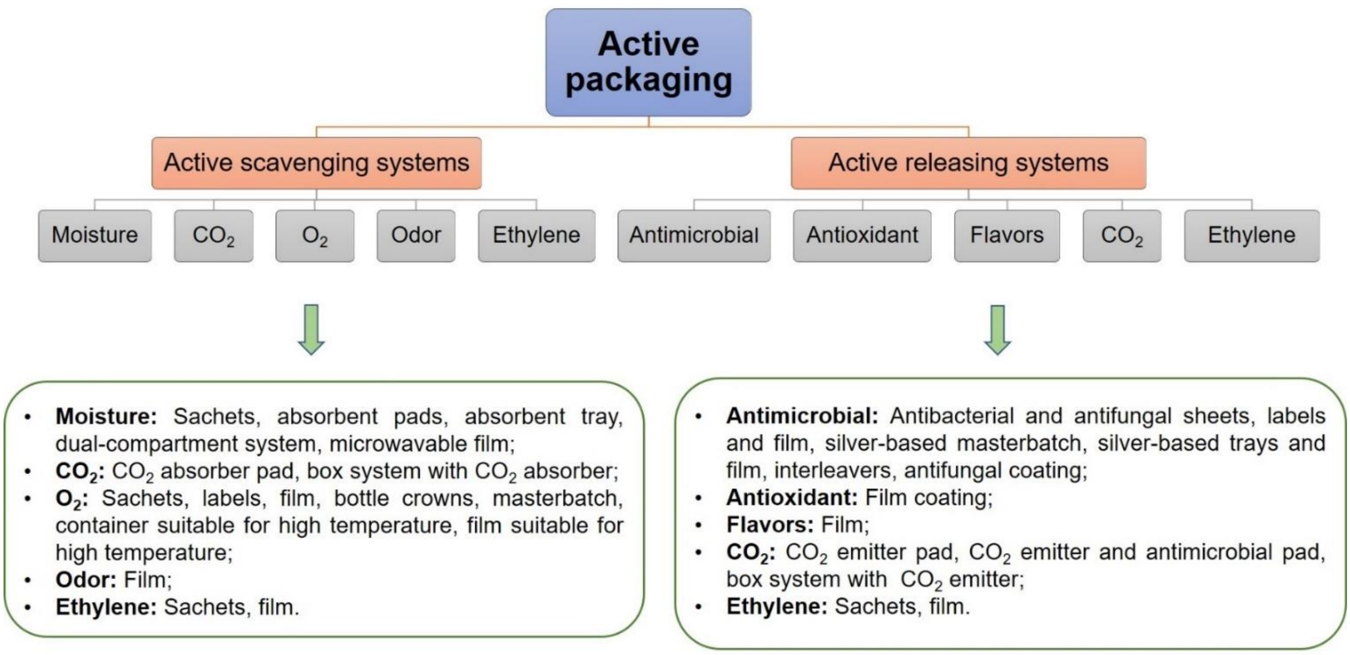


The approaches used in active packaging to gradually release active compounds include: chemical modification of polymers; development of multilayer films; use of cross-linking agents; molecular complexes and irradiation treatments; micro- or nano-encapsulation of active compounds; and nano-structured materials in the active packaging film matrix (Almasi et al. 2020). The chemical modification of polymers was initially proposed to guarantee improvements in mechanical, thermal and barrier properties, however, this technique proved to be efficient in controlled release systems, changing polymer-polymer reactions and promoting a longer contact time between bioactive compounds through laser treatment, plasma treatment and the use of chromic acid (Suganya et al. 2018). Multilayer films are composed of the barrier layer, matrix layer (containing the bioactive substance) and control layer, which controls the release of the active component into the food, and are normally produced by 2 different methods: coextruction and layer-by-layer deposition (Sharma et al., 2020b). The use of cross-linking agents refers, in practice, to the bonding of polymeric chains by covalent or non-covalent bonds, forming three-dimensional networks. The benefits associated with the controlled release system include decreased mobility of the polymeric chain and fixation of the bioactive compound (Benbettaïeb et al. 2019).

Irradiation treatment (such as ultraviolet radiation, gamma radiation and electron beam) is an innovative method that has been used to promote a more compact polymeric network with a high degree of crosslinking, resulting in the controlled release of active compounds (Almasi et al. 2020). Encapsulation is the most commonly applied method to protect the active compound and promote a more gradual release. The method consists of the imprisonment of the compound as the central material of a matrix known as wall material and is divided into 2 main groups: microcapsules (particles with a diameter of between 3 and 800 μm) and nanocapsules (particles with a diameter of between 10 and 1000 nm or 1 μm). The delivery systems used are classified based on the main wall materials used: lipid-based nanocarriers and distribution systems based on biopolymers (polysaccharides and proteins) (Ye & Chi, 2018). Active nanostructured materials are a fusion of nanocomposites, such as silver nanocomposites, and the active food packaging system, and they control the release of active nanocomposites through several mechanisms, such as: promoting strong interfacial interactions with reduced polymer chain mobility and acting as an immobilizing reservoir of active compounds, preventing an explosion-type diffusion in the initial stages of contact with food (Oleyaei et al. 2016).

The major methods reported through which active compounds are incorporated into polymeric materials are extrusion and the casting method (Chen et al. 2018). The casting method consists of solubilizing a polymer in a suitable solvent with the simultaneous incorporation of the active compound of interest; then, the solution is placed in a mold where the solvent is evaporated under controlled conditions, resulting in the formation of a plastic film of specific functionality (Nogueira et al. 2020). In extrusion, the active compound is incorporated with polymeric material melted by heat transfer, forming a mixture from which the films can be formed (Yepes et al. 2018). The casting method is widely employed on a laboratory scale, but it has practical and commercial limitations, i.e., small quantities of films are produced (Pelissari et al. 2019). Extrusion, although industrially viable, can present several limitations owing to the lack of thermal stability of many active compounds, which can be lost by degradation and evaporation during the heat transfer involved in the operations (Bastarrachea et al. 2011). Ensuring uniform distribution of the active compound and maintaining the mechanical properties of the films are additional challenges to the preparation of extruded active packaging materials (Bastarrachea et al. 2015). The main properties exhibited by active packaging are related to substances that absorb or produce bioactive compounds, as shown in Figure 5.

Figure 5. Main types of active packaging properties and their most common forms of presentation.



Source: Han et al. (2018); Wyrwa & Barska (2017); Yildirim et al. (2018).

Active films with antioxidant and antimicrobial properties have received greater attention owing to their ability to transport bioactive compounds and release them in a controlled way in food, helping minimize oxidation reactions, preserving microbiological and sensory aspects of food and, thus, extending the useful life of food products (Nieto et al. 2018). These compounds come from different natural matrices used in the preparation of active films and, in general, they originate naturally from the secondary metabolism of plants. Some of them exhibit powerful antimicrobial potential (essential oil of ginger, carvacrol, thymol), others act as potent antioxidants (Grape marc extract, yerba mate, rosemary, mint), and there is also the important class that acts as natural pigments (Anthocyanins, curcumin, betalain, carotenoids) (Arroyo et al. 2019; Bhargava et al. 2020).

These films have been widely used as packaging for foods in different segments, such as: fruits and vegetables (Kaewklin et al. 2018), extra virgin olive oil (Malherbi et al. 2019), cheeses (Bonilla & Sobral, 2018), meat products (Baek et al. 2019), breads (Priyadarshi et al. 2018b), roasted peanuts (Orsuwan & Sothornvit, 2018), among others.

Several studies have been performed with essential oils, natural extracts, and phenolic compounds, among other compounds, into biopolymer matrices, targeting antimicrobial (bactericide and fungicide) and antioxidant properties to produce active packaging. However, regarding virucide compounds into biopolymer matrices and how they behave in a food package or edible coating, studies are still scarce (Fabra et al. 2018; Randazzo et al. 2018). The interest in developing biodegradable and active packaging has been increasing and at a time when the world is experiencing a pandemic viral infection, the development of packaging with antiviral potential becomes essential. Antiviral products are designed through surface modification technology to destroy viruses, and antiviral activity is blocked by a thin film on the surface of the food (Pemmada et al. 2020). Naturally derived products and plant-based compounds such as green tea extract (Randazzo et al. 2017), grapefruit extract (Joshi et al. 2015a), *Hibiscus sabdariffa* extract (Joshi et al., 2015b), *Aloe vera* and Eriobotryae Folium extracts (NG et al. 2016), cinnamaldehyde (Fabra et al. 2016), carvacrol (Sánchez et al. 2015), thymol and zataria and oregano essential oils (Sánchez & Aznar, 2015) were studied, and they exhibited antiviral activity against enteric viruses (e.g., murine norovirus, hepatitis A virus, and feline calicivirus). Most antiviral coatings have been developed to be applied in packaging to prevent the spread of human enteric viruses (Chakhalian et al. 2020); however, owing to the emergence of the SARS-CoV-2 pandemic, greater attention should be paid to antiviral active packaging against coronaviruses.

SARS-CoV-2 is able to survive on the surface of packages (for example, it was detected for up to 72 h on plastic), which makes costumers concerned and raised interest in the development of polymers and biopolymers with antiviral activity (Doremalen et al. 2020; Olaimat et al. 2020). Although the risk of infection with COVID-19 when eating or handling food, including food packaging, is considered to be very low, the CDC (Center for Disease Control and Prevention) recommends washing one's hands with soap and water (at least 20 s) or use a hand sanitizer that contains at least 60% alcohol, after handling food packaging (CDC, 2020). According to the CDC (2020), no case of SARS-CoV-2 has been detected when touching food, food packaging, or shopping bags; however, it is essential to ensure good food safety practices to reduce common pathogenic diseases of food origin and to decrease the concern of the population regarding the virus remaining in food packaging.

Natural products such as herb extracts, phytocompounds, and essential oils have been demonstrated to be effective against SARS-CoV-1. The herbal extracts of *Lycoris radiata*, *Artemisia annua*, *Pyrrosia lingua*, and *Lindera aggregate* and its compound lycorine showed antiviral activity against SARS-CoV-1 in Vero cell-based assay in a virus-induced cytopathic effect screening (Li et al. 2005). The phytocompounds betulinic acid and savinin were evaluated against SARS-CoV-1, and the compounds behaved as competitive inhibitors of SARS-CoV-1 3CL protease (Wen et al. 2007). The essential oils from *Laurus nobilis* (IC_{50} of 120 μg/mL), *Thuja orientalis* (IC_{50} of 130 μg/mL), and *Juniperus oxycedrus* ssp. oxycedrus (IC_{50} of 270 μg/mL) exhibited activity against SARS-CoV-1 (Loizzo et al. 2008).

For SARS-CoV-2, Thuy et al. (2020), evaluated garlic essential oil by Docking simulation, and they found that the compounds in this essential oil inhibited the angiotensin-converting enzyme 2 (ACE2), causing the virus to lose the receptor in the host and, at the same time, the PDB6LU7 protein, which is the main protease of SARS-CoV-2. Thus, protein maturation is avoided and spread of the infectious process, indicating that this essential oil is a natural antivirus source. In another *in silico* study, Kulkarni et al. (2020) found that terpenes and phenylpropanoids - such as anethole, cinnamaldehyde, carvacrol, geraniol, cinnamyl acetate, L-4-terpineol, thymol and pulegone from essential oils extracted from plants belonging to the families *Lamiaceae*, *Lauraceae*, *Myrtaceae*, *Apiaceae*, *Geraniaceae,* and *Fabaceae* - were effective antiviral agents with the potential to inhibit the viral spike protein in SARS-CoV-2. Those studies have shown that there are diverse natural compounds that should be evaluated for the development of viable antiviral packaging, as well as applied in the medicine and pharmaceutical industry.

Recently, several studies have been published on the development and application of different active and biodegradable films. Table 1 shows some studies published in the last five years.

Table 1. Studies on the development of active films based on natural biodegradable components and their application as food packaging, in the last five years.

Active film composition	Valued property	Packaged food	Benefit	Reference
Chitosan and titanium dioxide nanocomposite	Antimicrobial and influence on the ripening process	Cherry tomato	The active film exhibited better ethylene photodegradation ability than chitosan film, favoring that rates of quality change in packaged tomatoes were slower during the entire storage. Besides that, the active film packaged samples did not show symptoms of fungal infection after storage for 13-14 days, due to effective antimicrobial activity against bacteria and fungi observed in preliminary studies of *in vitro* antimicrobial testing	(Kaewklin et al., 2018)
Chitosan and citric acid	Antioxidant	Green pepper	The film added with citric acid displayed enhanced antioxidant activity and was able to preserve for a longer period the green chilies packed as compared to those packed in neat chitosan films	(Priyadarshi et al., 2018a)

Chitosan with essential oils of cinnamon and ginger	Antioxidant and antimicrobial	Lean pork slices	The highest antioxidant and antimicrobial activities were observed in chitosan films incorporated with 1% of mixed essential oils and, in general, the active films were able to slow the total microbial growth, the increase in pH and lipid oxidation in the pork slices	(Y. Wang et al., 2016)
Chitosan and mango leaf extract	Antioxidant	Cashew nut	The antioxidant activity improved with the addition of mango leaf extract in the chitosan films, and the 5% mango leaf extract film was able to provide 56% more resistance to oxidation in cashew nuts than a commercial polyamide/ polyethylene film	(Rambabu et al., 2018)
Chitosan and grapefruit seed extract	Antimicrobial	Sliced loaf of bread	Chitosan films containing grapefruit seed extract (1.5% v/v) delayed the fungal growth on the bread surface by approximately 5-7 days when compared to the control film (low-density polyethylene) and the film containing pure chitosan,and the inhibition of fungal growth was proportional to the addition of grapefruit seed extract to the films	(Tan et al., 2015)

Chitosan and essential oil of ginger	Antimicrobial	Fish	The addiction of essential oil improved the antimicrobial activity of the chitosan film against foodborne pathogens, where the film with 0.3% essential oil showed the maximum antibacterial property against *Staphylococcus aureus* and *Escherichia coli.* The active film was efficient in extending the storage life of the fish, showing lower value of total volatile basic nitrogen and total mesophilic count when compared to the unwrapped control sample and aerobically packed sample in synthetic multilayer film of ethylene vinyl alcohol	(Remya et al., 2015)
Chitosan and apricot kernel essential oil	Antimicrobial	Bread	The active films showed antioxidant and antimicrobial activity, mainly against *E. coli* and *Bacillus subtilis* bacteria. The hydrogen peroxide and 2,2-difenil-1-picril-hidrazil elimination activity increased about 28 and 13%, respectively, of the pure chitosan films for chitosan films with maximum EO addition. The films also inhibited the growth of fungi in bread, thus increasing its shelf life	(Priyadarshi et al., 2018b)

Chitosan with microcapsules of grape seed extract and carvacrol	Antimicrobial	Atlantic salmon fillets	The active films showed lower values of total volatile basic nitrogen, pH, and luminosity after 7 days of storage in relation to control samples and control chitosan film. The CMF also showed lower values of mesophilic and psychrophilic bacteria and *Pseudomonas spp*, reaching the maximum limit allowed for the first two bacteria only on the 7th day of storage. The active films increase the shelf-life of refrigerated salmon to 4-7 days of storage due to the antimicrobial effect of the natural agents	(Alves et al., 2017)
Chitosan, carboxymethyl cellulose, and zinc oxide bionanocomposite	Antimicrobial	Egyptian soft white cheese	The active films showed good antimicrobial activity against total bacteria, fungi, yeasts, and coliforms in soft white cheese, in addition to show a significant impact on the studied pathogenic strains, presenting a inhibition zone that varied from 5 to 15 mm. Packaging films helped to extend the life of white soft cheese	(Youssef et al., 2016)

Chitosan, gelatin, essential oil of *Ziziphora clinopodioides*, cellulose nanoparticle, and pomegranate peel extract	Antimicrobial and influence on the ripening process (extension of shelf life)	Fresh peeled shrimp	All shrimps packed with chitosan and gelatin films had lower bacterial counts compared to the control films. The chitosan group treated with 1% Ziziphora essential oil + 1% pomegranate peel extract + 1% cellulose nanoparticle had the best antibacterial efficacy and the highest organoleptic scores after 11 days	(Mohebi & Shahbazi, 2016)
Zein, and chitosan nanoparticle with pomegranate peel extract	Antioxidant	Fresh pork meat	The active films showed a significant reduction in the growth of *Listeria monocytogenes* and extended the shelf life of packaged pork until the end of the designated storage period (14 days)	(Cui et al., 2020)
Carvacrol essential oil and agar	Antimicrobial	Fresh shitake mushroom	The film package was able to generate better quality preservation in terms of mushroom color, firmness, flavor score, and microbial counts after 6 days of storage at 10°C. The growth of *Saccharomyces cerevisiae* was completely inhibited by exposure to the active film and the growth of *Pseudomonas fluorescens* was slower, with smaller colony sizes in the medium after 10 days at 10°C	(H. J. Wang et al., 2016)

Zinc oxide nanoparticles and agar	Antimicrobial	Green grape	The green grapes wrapped in the active film with 2% and 4% of zinc oxide nanoparticles remained acceptable for 14 and 21-days, respectively, demonstrating that the efficiency of preservation of samples was directly proportional to the concentration of nanocomposite in the formulation	(Kumar et al., 2019)
Nanocomposite of banana flour and essential oil of garlic	Antioxidant and antimicrobial	Roasted peanuts	The active films showed antioxidant and antimicrobial capacity, especially the formulation with the addition of 1 mg mL^{-1} of garlic essential oil, which was able to inhibit the growth of *Aspergillus flavus* and was effective in preserving the quality of roasted peanuts having a similar effect to commercial plastic packaging.	(Orsuwan & Sothornvit, 2018)
Gelatin, chitosan and Chilean boldo extract	Antioxidant and antimicrobial	Slice cheese plate	Cheese samples packed in a film with Chilean boldo extract showed low coliform development and no growth of psychotropic microorganisms. All films demonstrated significant protection against oxidation compared to the control	(Bonilla & Sobral, 2018)

Cellulose acetate and pink pepper essential oil	Antimicrobial	Slice mozzarella cheese	The films added of essential oil presented antimicrobial activity against *L. monocytogenes* and *S. aureus* in all evaluated media (in a solid medium, broth, micro-atmosphere, and *in situ*). An affinity was observed between the nonpolar molecules of the essential oil and the lipid components of the cheese, in the *in situ* tests, suggesting that the developed films can be applied as active packaging, by direct contact	(Dannenberg et al., 2017)
Semolina flour and nanocomposites (zinc oxide and kaolin)	Antimicrobial	Diced mozzarella cheese	The films exhibited strong antimicrobial activity against *E. coli*, *S. aureus*, *Candida albicans*, and *Aspergillus niger*. The microbial growth in samples packed with the active film was less than 2.5 log CFU g^{-1} after 72 days of storage compared to the control, where the greatest effect of microbial inhibition was directly associated with the greater incorporation of zinc oxide in the formulation	(Jafarzadeh et al., 2018)

Fish skin gelatin and moringa oleifera leaf extract	Antioxidant and antimicrobial	Diced gouda cheese	The active film with the highest addition of extract in the formulation showed greater antioxidant activity. The film proved to be able to inhibit the microbial growth of *L. monocytogenes* in cheese samples. After 16 days of storage, the microbial count in cheese packed with active film decreased by 1.21 and 0.62 log CFU g^{-1} compared to the control samples and packed with the film without extract, respectively	(Lee et al., 2016)
Gelatin bionanocomposite, chitosan nanofiber, and zinc oxide nanoparticles	Antimicrobial	Chicken Fillet and white cheese pieces	The active film significantly decreased the growth of bacteria inoculated in chicken fillets and cheese samples. The changes in the pH values and color parameters were controlled during the storage time, in addition, the organoleptic characteristics in the chicken and cheese fillet samples packed with the active film were acceptable until the end of the storage (12 days)	(Amjadi et al., 2019)

Gelatin and durian leaf extract	Antioxidant	Durian fruit pulp	The films presented 17.6 times greater 2,2-difenil-1-picril-hidrazil elimination activity than the negative control sample. The film sample resulted in a three times lower oxidation rate of the palm oil compared to the negative control sample. This showed the efficiency of the gelatin film with the addition of 0.5% crude leaf extract in delaying the oxidation of the oil	(Kam et al., 2018)
Porcine plasma protein, chitosan, and encapsulated turmeric oil	Antioxidant	Riceberry rice	The films have maintained the quality of rice grains for at least 50 days and can be used as food packaging materials to maintain quality and extend the shelf life of food and agricultural products	(Samsalee & Sothornvit, 2020)
Whey protein isolate and *Lactobacillus casei* probiotic	Influence on the ripening process (extension of shelf life)	Thompson grapes	The films produced with the addition of *Lactobacillus casei* presented adequate probiotic counts during 14 days of storage, with a positive effect on the ripening of the fruits evidenced by the lower total soluble solids (TSS) content, being an alternative to prolong the ripening of the product and bring benefits to the health of the product consumer	(Dianin et al., 2019)

Cassava starch, oregano essential oil, and pumpkin residue extract	Antioxidant and antimicrobial	Ground beef	The active films demonstrated antioxidant activity *in vitro*, antimicrobial activity against coliforms, mesophilic bacteria, and *Salmonella spp*, and contributed to the maintenance of low pH values until the sixth day of storage	(Caetano et al., 2017)
Protein from dry residues of distilled grains and tea extract (green, oolong, and black)	Antioxidant	Diced pork	During storage, the pork meat involved with the active films had less lipid oxidation when compared to the control films. Among the tea extract, the film with green tea extract showed greater antioxidant activity	(Yang et al., 2016)
Black-eyed bean starch and Chilean blackberry extract	Antioxidant	Diced salmon	The Chilean blackberry extract at 20% concentration, led to an increase in the activity against the radicals 2,2-azinobis(3-ethylbenzothiazoline-6-sulfonic acid) and 2,2-difenil-1-picril-hidrazil of 88.46% and 42.39% respectively, and the active film delayed the lipid oxidation in salmon samples throughout the storage at 4° C	(Baek et al., 2019)

Corn starch, gelatin, and guabiroba pulp	Antioxidant	Extra virgin olive oil	The extra virgin olive oil samples stored in the active films and control did not exceed the maximum values of the acidity and peroxide index allowed by Brazilian legislation. The addition of fruit pulp to the film formulation did not affect the oxidative stability of extra virgin olive oil	(Malherbi et al., 2019)
Corn starch and microencapsulated anthocyanin extracted from grape marc	Antioxidant	Olive oil	The active film was able to maintain the quality attributes of extra virgin olive oil, which showed good oxidation stability under accelerated thermal and photo-oxidative degradation environments. Olive oil showed peroxide, acidity, and K_{232} values below the limit established by Codex until the 8th day of storage when packed with the active film, and high degradation rates on the 4th day when packed in polypropylene	(Stoll et al., 2017)

Cassava starch, polyvinyl alcohol, sodium alginate, and essential oils of lemongrass and copaiba	Antimicrobial	Minimally processed lettuce	Biodegradable films with copaiba oil showed increasing counts of molds and yeasts after the 8th day of storage, but lower in comparison with the other treatments, indicating that this film presented greater control of these microorganisms. The mesophile count increased significantly for all treatments at the end of storage. The addition of essential oil did not exercise control over the total coliform development	(Brandelero et al., 2016)
Pinhão starch and feijoa peel flour	Antioxidant and antimicrobial	Apple	The greater the addition of flour in the films, the greater the concentration of antioxidant compounds and the antimicrobial activity against *E. coli*, *Salmonella typhimurium*, and *Pseudomonas aeruginosa*. The active films and the control film did not show significant differences in the weight loss of the apples, and after the 5th day of storage, the weight of the packed apples remained constant, unlike the unpacked apples, which lost weight until the 15th day of storage	(Sganzerla et al., 2020)

Furcellaria, isolated whey protein and extracts of yerba mate and white tea	Antioxidant and antimicrobial	Fresh rennet cheese	The cheese samples wrapped with the active film were characterized by a lower yeast and fungus count than the cheese packed with the control, in addition, the active packaging was also able to inhibit the growth of coliform bacteria, increasing the shelf life of the cheese. The active film samples positively impacted the organoleptic quality and the consistency of the packaged cheese	(Kubica et al., 2019)
Semi-refined carrageenan and germinated fenugreek seed extract	Antimicrobial	Chicken breast	The films showed a significant increase in the phenolic content and antioxidant activity proportional to the increase in the concentration of germinated fenugreek seed extract. In addition, the active films showed a significant control on the total bacterial count on the surface of chicken breast meat during the 7 days of cold storage, resulting in a microbiological shelf-life extension of 6 days when compared to the control	(Farhan & Hani, 2020)

Alginate and pineapple peel active compounds	Antioxidant	Beef meat	The films containing pineapple peel active compounds exhibited greater antioxidant activity than the control, showing great potential to delay lipid oxidation in meat samples. The active films also performed well in inhibiting the growth of aerobic mesophilic bacteria during the 5 days of storage under refrigeration	(Lourenço et al., 2020)
Papaya puree, ascorbic acid, and Moringa oleifera extract	Antioxidant	Minimally processed pear	The active films showed satisfactory antioxidant activity, proposed as an alternative method for extending the shelf life of foods susceptible to oxidation, such as pears. Ascorbic acid was primarily responsible for this beneficial effect, also positively influencing sensory acceptance	(Rodríguez et al., 2020)

Rye starch, sorbitol, and rosehip extract	Antioxidant	Chicken breast	The active films showed great antioxidant potential, with high activities of elimination of radicals 2,2-azinobis(3-ethylbenzothiazoline-6-sulfonic acid) and 2,2-difenil-1-picril-hidrazil (96.87% and 80.22%, respectively). Chicken samples packaged with the active film demonstrated lower values of peroxide and substances reactive to thiobarbituric acid than films without rosehip extract and the control	(Go & Song, 2019)
Gelatin with chitosan nanocomplexes and anthocyanins	Antioxidant	Olive oil	The active films containing anthocyanins significantly delayed the oxidative deterioration of olive oil (21.2 meq O_2 Kg^{-1} of peroxide value at the 56th day), when compared to the other films, this bioactivity was attributed to the antioxidant power of anthocyanins	(Wang et al., 2019)
Triticale flour with bacteriocin-like substance	Antimicrobial	White semi-firm cheese slices	Active films maintain antimicrobial activity until 45 days of evaluation. This package can reduce the *Listeria spp.* count by 3 logs compared to the control after 15 days of storage	(Salvucci et al., 2019)

Alginate-oleic acid-based coatings containing green tea extract	Antiviral	Strawberries and raspberries	The infectivity of the murine norovirus (MNV), and hepatitis A virus (HAV) in strawberries after the coating treatments was reduced by 1.5-2 log during the 4-days storage period at 10°C as compared to the controls	(Falcó et al., 2018)
Lipids, green tea extract, and grape seed extract added to alginate films	Antiviral and antioxidant	-	The films exhibited significant antiviral activity against MNV and HAV since ~2 log reduction was recorded for the 0.75g extract/g alginate in the film and the incorporation of phenolic compounds imparted antioxidant capacity to the active films	(Fabra et al., 2018)
Film-forming dispersions based on κ−, ι− and λ− carrageenans, containing green tea extract	Antiviral	Blueberries and raspberries	All the coatings were effective in extending the shelf life of the fruits under refrigeration, preserving their firmness to a greater extent and promoting better appearance, in addition to better preservation of the fruits in terms of antiviral infectivity at refrigerated and ambient conditions, which was intensified by the presence of green tea extract	(Falcó et al., 2019)

Agar and agar/alginate coating containing *Larrea nitida* extract	Antiviral	Blueberries	The coatings of agar and *Larrea nitida* extract were able to reduce the infectivity of MNV below the limit of detection after over-night incubation at 25°C and after 4 days at 10°C storage	(Moreno et al., 2020)
Persian gum and gelatin coating, containing allyl isothiocyanate	Antiviral	Blueberries	MNV was decreased by 3.25 log and 3.00 log for 0.1% and 0.5% of allyl isothiocyanate, respectively after overnight incubation at 37 °C	(Sharif et al., 2021)

Legend: MNV - murine norovirus; HAV - hepatitis A virus

There is a growing tendency in the last years of using films based on natural biodegradable components in several food matrices, and they are also employed as probiotic and bioactive compound carriers, providing beneficial activity on the food product. In addition, owing to their attractive characteristics of color, flavor, and aroma, they can be used to improve the sensory characteristics of food, and consumed as an integral part of it, e.g., in snacks, sushi wraps, soluble drinks, or instant soups. However, there are still challenges to overcome in order to guarantee broader applicability in different matrices: for example, increasing barrier properties, since most films based on natural compounds have great hydrophilicity and high-water vapor permeability. Studies about these characteristics serve as guidelines for application of films; for this reason, there needs to be further research on the development, characterization, and application of biodegradable active films for food packaging.

7. From agro-industrial waste to active food packaging: opportunities and challenges in the circular economy

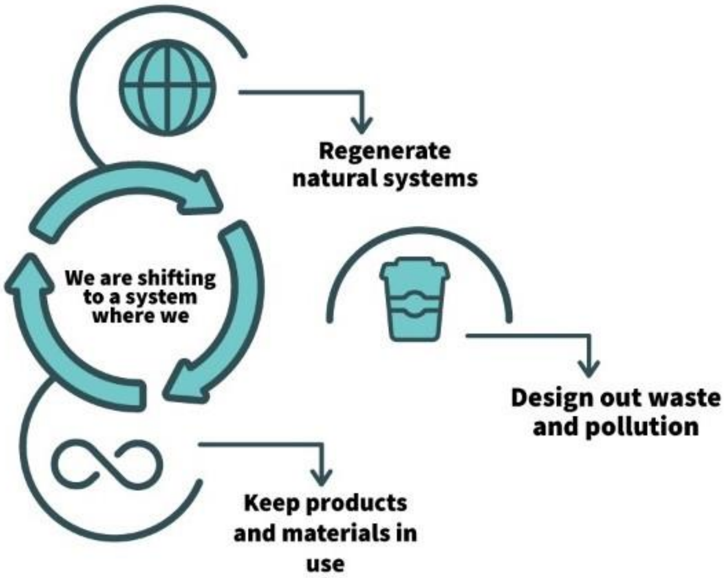
The use of food packaging, despite all the fundamental benefits involved, is a major concern for the preservation of the environment, given the high production volume and intense discard, mainly associated with the short usage time, which makes waste management even more complex (Geueke et al. 2018).

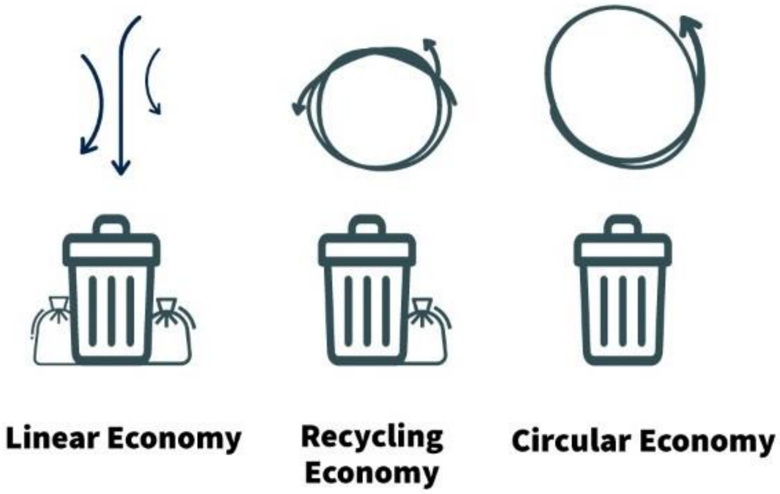
A few years ago, transforming materials that would be discarded into something new was practically the exclusive work of artists and artisans. Currently, several types of materials are reused in upcycling, such as plastic and paper, reducing not only the demand for new raw materials for product manufacturing, but also the consumption of energy and water involved in this process. The goal is to reach a circle of production and consumption in which waste is seen as input, not as waste. This is the idea of a circular economy, based on 3 basic principles: “To reduce, to reuse, and to recycle”. Reduction aims to minimize the use of raw materials and waste production; the principle of reuse is based on the repeated use of products for their intended purpose, and recycling is mainly used to save energy, resources and emissions, in addition to reducing the environmental impact caused by the use of materials. It is evident that this model works by remodeling the form of production of economic goods, with attention to sustainable development (Abre, 2019; Geueke et al. 2018).

The circular economy encloses different layers encompassing resource scarcity, environmental, and social impact and economic benefit, the last one being the point that commonly has aroused the most interest (Niero & Hauschild, 2017). It is known that about 39% of the European demand for plastics is for packaging production, which prompted the European Union’s (EU) initiative to put in place a general action plan for the circular economy, in which one of the guidelines presents legislative proposals around a common goal: recycling 75% of packaging waste by 2030 (Plastics Europe Associaton of Plastics Manufacturers, 2020).

Most disposable packaging has a relatively short life cycle, i.e., less than 1 year, as it is disposed of immediately after use, automatically entering the waste stream. According to Clark (2020), workers buy an average of 4 items packed for lunch, which will mostly be used only once, generating about 10.7 billion separate items of garbage per year. A report produced by the EU on the "Single Use Plastics Directive" showed that after a short first use cycle, the value of plastic packaging material is lost by 95%. We seek to emphasize the dichotomy between a linear flow of production, which is established with the "produce, consume and dispose" system, based on the idea that economic growth can be based on the abundance of resources and the unlimited elimination of waste; and the circular flow of production, in which waste is seen again as a raw material through reuse, repair, reconditioning and recycling of existing materials, adopting the idea that industrial systems function as natural ecosystems (Niero & Hauschild, 2017). Figure 6 shows the difference between linear and circular economy.

Figure 6. Objectives of the circular economy and difference with the linear flow of production.





Sources: Ellen MacArthur Foundation (2021); AX Foundation (2020).

Even if conventional plastics can be recycled, research and development for biodegradable materials is still important, with special concern to those materials that will be in direct contact with food, i.e., primary food packaging. It is of great importance to assess the safety of conventional recycled plastic packaging as recycling can increase the levels of hazardous chemicals in packaging and food (after migration). Plastics that are recyclable are those made from thermoplastic polymers such as polyethylene terephthalate (PET), polypropylene (PP), high (HDPE) and low (LDPE) density polyethylene, polystyrene (PS) and polyvinyl chloride (PVC). These types of plastics can be mechanically recycled, and partial degradation of the polymer skeletons occurs through heating, resulting in modified mechanical and optical properties, such as increased fragility and opacity (Geueke et al. 2018; Groh et al. 2019).

Thus, attention should be paid to some problems during application of recycled plastics for food packaging purposes, as they can present various chemical contaminants, including odor and taste compounds from cross-contamination; oligomers and monomers derived from polymeric degradation during recycling; additives and their degradation products arising from degradation reactions or cross-contamination; inorganic elements of environmental origin. In other words, recycling of these plastics waste for use as food packaging can often result in a product with lower quality i.e., with reduced chemical safety standards (downcycling), which is a major food safety problem (Geueke et al. 2018; Horodytska et al. 2020).

If, on the one hand, we may have contaminants through the use of recycled plastics, on the other hand, not using them generates a large accumulation of waste, leading to negative impact on various ecosystems. This is why it is essential to invest in biodegradable polymers that can be used as active bio-based packaging, helping to conserve packaged foods and acting on two key points: increasing the shelf life of food products and taking advantage of residues that would be discarded otherwise (Geyer et al. 2017).

Moreover, another factor of great environmental importance is to pay attention to agroindustrial waste as a potential source of renewable and environmentally friendly primary material, which can be used as a raw material in innovative processes. This initiative might be favorable to the reduction of postharvest loss or waste and also help to offer a solution to pollution problems related to the disposal of conventional plastics (Matheus et al. 2020). Several studies have been explored the use of agroindustrial residues for biodegradable food packaging (Brito et al. 2019; Tran et al. 2020), including combining the active character in these packaging (Sganzerla et al. 2020; Szabo et al. 2020).

Vegetable residues have great potential for use owing to the variety of compounds present in the food matrix, such as carbohydrates, proteins, lipids, fibers, vitamins, minerals, in addition to several bioactive compounds (Brito et al. 2020;

Georganas et al. 2020). A great deal of research is focused on finding a solution and overcoming technical problems arising from the use of this type of bio-based material, especially because of the great variability of the available raw material and the narrow processing window, which impairs large-scale production and diffusion of packaging on the market. In addition, there is still a lack of tools to help adapt the packaging to the needs of the products (Guillard et al. 2018).

There are some complex challenges to the development of food packaging within the concept of circular economy, as it affects a wide range of interconnected sectors and requires a plurality of approaches. Innovation in the food market requires a multi-scale, multidisciplinary and multifactorial approach, involving initiatives by politicians, industries, researchers and consumers, who play a relevant role in the sustainability of the food chain. In addition, actions that contribute to the general reduction of food losses and the development of more sustainable plastics have great potential (de la Caba et al. 2019; Geueke et al. 2018).

The principles of the circular economy are being increasingly applied by the largest companies in the world, from different sectors and value chains (Ellen Macarthur Foundation, 2021). The objective of the company Unilever, for example, is not only to use less packaging, but also to design it in a way that it can be reused, recycled or composted, i.e., causing a reduction of plastic in the shared environment, contributing to the UN’s Sustainable Development Goal (SDG) on Sustainable Consumption and Production (SDG 12) (Unilever, 2020). Ambev is another great brand focused on driving progress towards the UN’s SDGs. Between 2012 and 2019, the company removed more than 200 thousand tons of packaging materials from Brazil through the use of recycled materials, in addition to reaching over 99% internal recycling rate in its breweries. As a target for 2025, the company aims to make all packaging returnable or made mostly of recycled content (Ambev, 2020). There are other companies aligned with these objectives, including McDonald's, Kraft-Heinz, PepsiCo and Coca-Cola. (Boz et al. 2020).

The incorporation of circular economy precepts in the political guidelines of different countries has the fundamental role of driving and strengthening more sustainable development, once initiatives coming only from large industries may make it difficult to achieve results on a large scale (Ellen Macarthur Foundation, 2021).

In this sense, the European Union has stood out in recent years as one of the regions that most often applies policies for sustainable development, including waste reduction and recycling. Controversially, it still presents a large consumption of resources, and European countries differ with regard to recycling rate (Ferronato et al. 2019). In 2016, some countries presented high recycling rates, such as Belgium (81.9 %), Germany and Spain (around 70.0 %), Ireland, Italy, Austria, and France (between 66.0 and 67.0 %), while others, such as Malta and Hungary, presented lower rates, 39.7 % and 49.7 %, respectively (EEA, 2019).

In addition to European countries, the United States has also been working to boost the circular economy, aiming at higher recycling rates (Meys et al. 2020). Among Asian countries, China is seeking to increase recycling rates (Meys et al. 2020), and Japan is trying to reduce the use of single-use plastics and encourage substitutes for conventional plastics (Jang et al. 2020).

Usually, it can be seen that developing countries have greater difficulties in implementing policies based on the principles of the circular economy owing to numerous factors, such as less advanced technologies, lower financial resources, lower public awareness, and weaker political will (Ezeudu et al. 2020; Ferronato et al. 2019). It is important to take advantage of this current moment in the world, shaken by the COVID-19 pandemic and in search of a resilient economic recovery, to redefine the system and develop new and more environmentally friendly mechanisms (Ellen Macarthur Foundation, 2021).

8. Perspectives

Changes in consumer demand and lifestyle, trends in industrial production, and retail practices are the main driving forces behind the evolution of new food packaging technologies. With the constant increase in environmental issues related to the accumulation of plastic waste, active bio-based packaging has been seen as a viable alternative to circumvent this problem, considering not only the application of this material in a circular economy system but also the improvement of quality and safety of packaged foods, owing to the presence of intrinsic bioactive compounds. However, the main barrier to the absorption of the market is attributed to technical bottlenecks related to the functional specificities and production of bio-based materials, which are quite different from those of petrochemical plastics, namely thermal instability, high permeability to oxygen and water vapor, difficult thermal sealing and high production cost. As a result, different formulations and the use of reinforcement nanoparticles are being extensively explored to overcome these obstacles and produce bioplastics with good mechanical and barrier properties in an economically feasible form. In addition, one needs to consider other aspects for the development of new plastics, with greater responsibility for the impact caused on different ecosystems, moving towards the use of active packaging for food, based on the use of functional, innovative, and sustainable materials.

9. Conclusions

There is a clear trend towards an increase in the consumption of plastic packaging worldwide as a result of the COVID-19 crisis. There has never been a more urgent need to explore active biodegradable materials for food packaging, among other applications. The use of agro-industrial residues as a source of added value to produce bioplastics can help to reduce white pollution and generate additional income. Recent studies have focused on overcoming the technological gaps and the high costs associated with alternative natural materials, particularly regarding the difficulties related to production on an industrial scale, reduced barrier properties, and the guarantee of the stability of the bioactive compounds on active packages. In addition, advances in nanotechnology have great potential to improve biodegradable and active packaging. It is necessary to consider how the current model of production is changing with the emergence of the circular economy model to the detriment of the linear economy and the insertion of the innovations proposed in this context. Finally, it is believed that research and development must be symbiosis with government incentives, multidisciplinary research, industries, and consumers. Awareness-raising campaigns are necessary as a long-term strategy to increase consumers' behavioral attitudes and social responsiveness to tackle the threat of plastic pollution.

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AUTHOR CONTRIBUTIONS

Barone, AS and Fai, AEC designed the study; Barone, AS; Matheus, JRV and Souza TSP wrote the paper with input from all authors; Fai, AEC and Moreira, RFA provided critical revision of the article; Fai, AEC supervised the project.

CONFLICT OF INTEREST

The authors declare no conflict of interest.

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CAPÍTULO II

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Development of biodegradable food packaging in the context of COVID-19: sustainability more urgent than ever

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ABSTRACT

The indiscriminate overconsumption of non-biodegradable packaging, particularly single-use food packaging, resulting from COVID-19 is exacerbating an age-old environmental problem. This is due to changes in consumer behavior that is concerned with safety issues when buying food, which has led to a preference for purchase in delivery and take-out. In addition, health department agencies indicate the use of disposable products to prevent contamination (e.g., disposable foods; and pre-packaged foods). To date, there is no evidence of SARS-CoV-2 contamination through food ingestion, however, COVID-19 has raised awareness about packaging for hygienic-sanitary protection of food. To minimize the impacts related to the increase in the consumption of plastics, there must be a greater investment in research for the development of biodegradable packaging and also a change in the type of economy adopted nowadays - linear economy, to the circular economy. Since biodegradable packaging brings less impact to the environment, and the circular economy presents as fundamentals reduce, reuse, and recycle. Thus, packaging material types and the way we deal with waste must be adapted urgently.
Keywords: biodegradable food packaging, circular economy, COVID-19 impacts, one single used plastic

1. Introduction

Plastic manufacturing has surpassed most other man-made materials, with over 90% being petroleum-based and non-biodegradable. Packaging, particularly food packaging, largely derives from plastic and is a leading contributor to urban waste (Zhao et al. 2020). This scenario has become a transboundary threat to natural ecosystems and has been aggravated by the overconsumption of single-use food packaging and plastic grocery bags due to the COVID-19 pandemic (Silva et al. 2021).

The paradox that emerges lies in the possible association between COVID-19 and environmental issues. Environmental management plays a prominent role in the exposure and spread of infectious diseases, mitigation of pollutants, and regulation of climatic factors. Pollutants and some viruses - such as SARS-CoV-2 - cause negative immune responses and share similar mechanisms of action. Therefore, they may play an additive and reinforcing role in viral diseases like COVID-19 and must be considered as part of the approach to prevent future diseases (Espejo et al. 2020).

As stated by Albert Einstein, “We can't solve problems by using the same kind of thinking we used when we created them”. Thus, urgent reflection upon achievements, current challenges, and perspectives of biobased and biodegradable food packaging materials is necessary to contribute to a greener future and global sustainable recovery from the pandemic.

This study presents insights into the current development of biodegradable food packaging and future trends in the context of COVID-19.

2. Eating habits and lifestyle changes due to the pandemic leads to more single-use plastic: an old problem, a new emergency

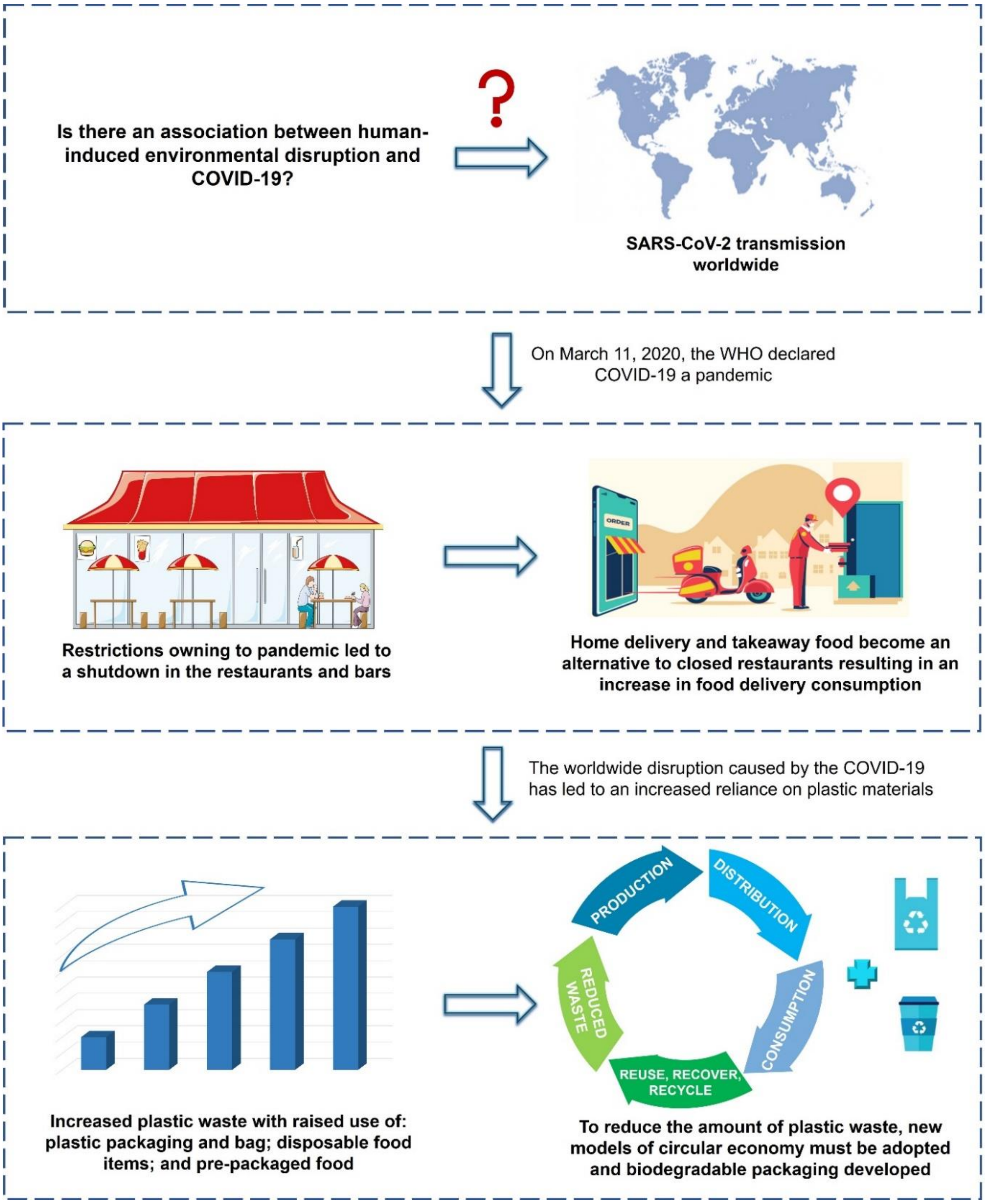
The demand for food delivery and takeout has potentially increased since the COVID-19 pandemic imposed major changes in human activities. These services promote easier access to prepared meals and allow food suppliers to maintain their activities (Li et al. 2020). Owing to safety concerns when going out to supermarkets or restaurants, there has been a shift in consumer preferences towards e-commerce purchases (Parashar and Hait 2021).

As a result of this change in behavior, the demand for single-use plastics has increased. It is expected that this demand has risen by 40% in packaging and 17% in other applications (e.g., medical uses) (Silva et al. 2021). This increased requirement is mainly due to food delivery and takeout packaging, as the Centers for Disease Control and Prevention (CDC) consider that the lowest risk of COVID-19 spreading in food service is when it is limited to drive-through, delivery, takeout, and curb-side pick-up; the CDC also recommends the use of disposable food service items in bars and restaurants (CDC 2020a); and pre-packed foods in grocery stores, owing to safety concerns related to shopping in supermarkets during COVID-19 (Prata et al. 2020) (Fig. 1). (Filho et al. (2021) assessed the increase in consumption and subsequent changes in the quantities of waste produced since the COVID-19 pandemic. The study was carried out on 204 consumers from 23 countries. They found that 45-48% of the interviewees noticed a growth in their consumption of packaged food, fresh food, and food delivery; and an increase of 53% in plastic packaging. Another interesting finding was that consumers have been more critical of food producing companies and many calls for the development of products that use less packaging or increase the recyclability of the packaging they use.

Concerns regarding reusable plastics as carriers of COVID-19 have led consumers to seek more hygienic and disposable options (Prata et al. 2020; Yates et al. 2021). For instance, consumers have preferred fresh-food packaged in plastic containers, and the use of single-use food packaging to prevent food contamination (Silva et al. 2021; Yates et al. 2021). Nevertheless, there is no evidence that single-use packaging contributes more or less than reusable packaging to the spreading of coronavirus (Prata et al. 2020). In addition, according to the CDC, there is currently no evidence that the SARS-CoV-2 virus can be transmitted through food or food packaging (CDC 2020b). As COVID-19 increased awareness of packaging for hygienic-sanitary protection of food, the plastic industry voiced concerns regarding food safety, hygiene, and cross-contamination when using reusable containers and bags during the pandemic, leading to increased use of single-use plastics (Silva et al. 2021).

It is expected that in post-pandemic life, e-commerce purchases will continue to grow which will contribute to a substantial rise in packaging material, thus resulting in further problems regarding treatment and recycling (Gorrasi et al. 2021). Thus, extensive use and improper disposal of plastic waste will quickly become a global issue (Silva et al. 2021). To avoid this situation, there must be an immediate transition to new models of a circular economy and a greater commitment should be given to the development of packaging with new materials, such as biodegradable materials (Gorrasi et al. 2021) (Fig. 1).

Figure 1. Changes owing to the pandemic that occurred in the food service that led to a rise in plastic waste.



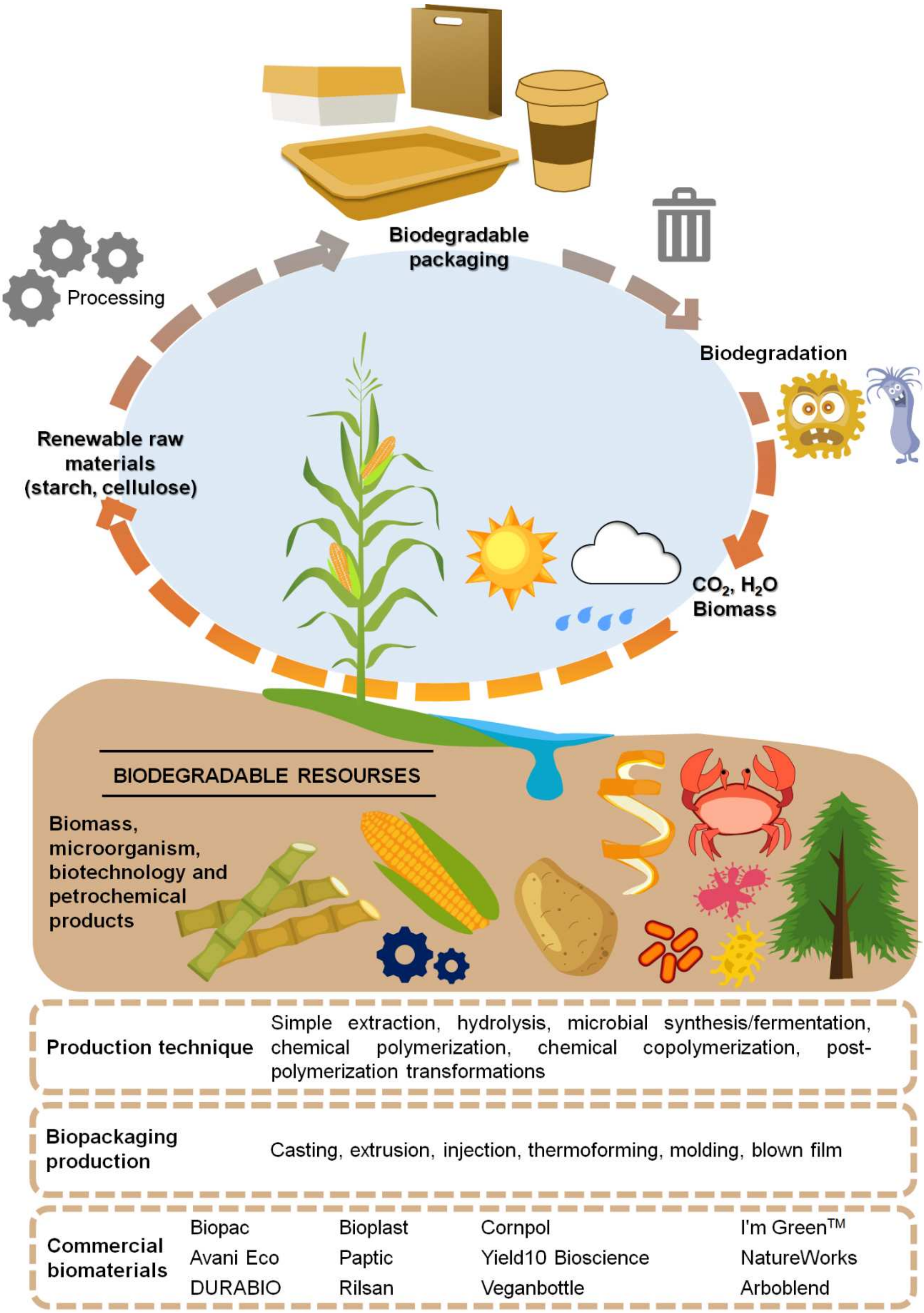
3. Development of biodegradable food packaging: recent achievements and current challenges

The packaging industry is increasingly looking into bio alternatives, particularly biodegradable versions, to enhance its sustainability. The market for biodegradable packaging has been driven by the need to develop innovative approaches that meet growing demands and reduce the adverse effects of traditional materials, particularly plastic. The market for bioplastic packaging presents growing demand and was valued at US $ 8.3 billion in 2019, with the expectation that by 2027 it will reach US $ 27.39 billion (Grand View Research 2020). The main brands and companies operating in the market for biodegradable materials include Biopac (Biopac Ltd, United Kingdom), Bioplast (Biotec GmbH, Germany), I'm Green (Brasken, Brazil), Avani Eco (Indonesia), Paptic (Netherlands), Yield10 Bioscience (USA), Durabio (Mitsubishi Chemical, Japan), Rilsan (Arkena, France), Veganbottle (Lyspackaging, France), Ecovio L (BASF, Germany), Bionic Bio-Fan (ZIEHL-ABEGG, Germany).

Unlike conventional plastic packaging, biodegradable packaging is made from biodegradable polymers, derived from bio-renewable sources that allow the packaging to be biodegraded or to be completely composted by hydrolytic or enzymatic cleavage of the polymer bonds (Sangroniz et al. 2019).

According to the source and synthesis process, biodegradable polymers can be classified into different types (Fig. 2): polymers obtained directly from biomass, such as proteins, lipids, and polysaccharides; polymers resulting from the metabolism of microorganisms, such as poly-hydroxyalkanoates and poly-hydroxybutyrate; polymers from biotechnological processes (such as polylactic acid); and polymers derived from petrochemical products, such as polycaprolactones and polyesteramides (Zhong et al. 2020).

Figure 2. Biodegradable packaging material – types and technological approaches.



Materials such as proteins (collagen, gluten, and zeins), lipids (beeswax, carnauba, and free fatty acids), and polysaccharides (starch, cellulose, chitosan, alginates, and their derivatives) have historically been the most evaluated raw materials for the processing of biodegradable packaging. In addition to being versatile and ecologically sustainable, these materials have properties that make them potential substitutes for some polymers of petrochemical origin (Nilsen-Nygaard et al. 2021). In general, packaging materials must be resistant to breakage and abrasion, to protect and reinforce the structure of the products, and they must also be flexible, to adapt to possible deformations without breaking. However, packaging made up of only one type of macromolecule has fragile mechanical properties, becoming brittle mainly as a result of humidity and temperature changes (Mohamed et al. 2020).

Several strategies have been researched in order to circumvent such limitations, such as the addition of plasticizers such as polyalcohols, e.g., glycerol, propylene glycol, and unsaturated or saturated fatty acids, caprylic and stearic acids, responsible for elevating plasticity and generating a material with greater elongation and flexibility (Zhong et al. 2020). Other alternatives include the use of physical, chemical, and biochemical modifications, such as thermal, chemical, and enzymatic cross-linking, ionizing irradiation, and the use of mixtures, such as the incorporation of antimicrobial or antioxidant compounds and nanoparticles. The latter form bionanocomposites, non-toxic, high-performance, light, and ecological materials, capable of improving thermal, mechanical, and barrier properties, without jeopardizing the biodegradable characteristics of the packaging. Nano clay, montmorillonite, zinc oxide, titanium dioxide, and silver nanoparticles are among those most commonly used (Youssef and El-Sayed 2018).

In general, studies on biodegradable packaging have focused on finding new sources and improving production processes in order to guarantee the availability, accessibility, and delivery of food, as well as contributing to the maintenance of a healthy ecosystem and environment (Petkoska et al. 2021). From this perspective, regulation is necessary for the safe and efficient manufacture and commercialization of packaging produced with biodegradable materials.

4. Perspectives and future directions

Governments, companies, and civil society have been implementing several strategies to mitigate the problems associated with the widespread use of disposable plastics from non-renewable and non-biodegradable sources. In the existing economic system (linear economy), food packaging is usually designed for single-use and discarded after short periods of time. To overcome the environmental impacts of packaging waste, the circular economy has been proposed. The three fundamental principles of the circular economy are to reduce, reuse, and recycle. It aims to remodel the production of economic goods, focusing on sustainable development. The main drivers of this transformation are the increased awareness of consumers about environmental issues; greater commitment from companies to develop projects; campaigns involving more sustainable practices for food packaging; and greater government awareness measures to support and guide these changes (Geueke et al. 2018), since initiatives from large industries alone can make it difficult to achieve large-scale results.

In addition, new compostable or biodegradable materials have been used to replace conventional plastics (Ellen MacArthur Foundation 2021). During the pandemic, a number of big companies launched new prototypes of environmentally friendly packaging, such as paper packaging for yogurt (Vigor 2021), paper packaging and fully recyclable plastic for soda (Coca-Cola 2018), and recyclable plastic packaging for chocolates (Nestlé 2021).

If on one hand, the pandemic has contributed to the increase in the use and disposal of conventional plastics, on the other, it has brought attention to new opportunities for the food packaging area (Fig. 3). Finally, another relevant aspect highlighted by the pandemic in the context of food packaging, is that companies must take into account the psychological needs of consumers from a hygienic-sanitary point of view. Some strategies have been adopted for this purpose, such as tamper-proof seals and airtight caps (Packhelp 2021). Another example of new packaging with emotional and market appeal motivated by the pandemic is a flexible eco-friendly film developed by Terphane, which is said to be effective against SARS-CoV-2 and focuses primarily on food delivery packaging (Abre 2021). The pandemic has further evidenced the demand for a sustainable and resilient food system. To improve the sustainability of food packaging and reduce environmental impacts, there must be a shift from the current linear economy to the circular economy model in addition to increased research on alternative materials to replace single-use conventional plastics such as biodegradable materials from renewable sources and introduce active and intelligent packaging by using natural and environmentally friendly bioactive molecules.

Figure 3. Expectations for new market trends in the area of food packaging during/post-pandemic.

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CAPÍTULO III

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Active antioxidant and aromatic films based on persimmon (*Diospyros kaki* L.) and orange peel flour (*Citrus sinensis*)

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ABSTRACT

Active antioxidant and aromatic films based on persimmon (*Diospyros kaki* L.), which was added to orange peel flour (*Citrus sinensis*) and glycerol, were developed and characterized. The films showed high values of water vapor permeability (7.09 – 12.61 × 10^{-6} g h^{-1} m^{-1} Pa^{-1}) and solubility in water (WS) (45.27 – 75.80%) and 0.2 – 1, 71 MPa, 10.24 – 15.84% and 1.8 – 15.32 MPa of tensile strength (TS), elongation at break (EB) and Young's modulus (YM), respectively. The addition of orange peel flour increased the TS, while the incorporation of glycerol increased the EB and WS of the films. The films were largely red and yellow, indicating a natural tendency to maintain the color of the persimmon pulp. Thermogravimetric analysis showed two important degradation steps related to glycerol and pectin (130°C - 230°C) and polymer structure decomposition (above 300°C). Scanning electron microscopy revealed a homogeneous and compact structure of the films, which obtained a rougher texture with the addition of glycerol. The addition of orange peel flour to the film-forming solution proportionally increased the total phenolic content and antioxidant capacity of the persimmon puree films. A total of 71 volatile compounds belonging to different chemical classes were identified, including limonene, β-ionone and vacenic acid, which influence the overall aroma, as they were detected above their odor thresholds, and the functional properties of these films. This study indicates the potential of persimmon films incorporated with orange peel flour as active antioxidant and aromatic primary packaging materials for foods in a circular economy context. It also indicates the potential use of persimmon and orange peel flour films as active and biodegradable packaging.
Keywords: Food waste valorization; Bioeconomy; Composite films; Biodegradable films; Barrier and mechanical properties; Bioactive compounds

1. Introdution

Technological advances and growing concern for the environment have stimulated the development of biodegradable plastics from renewable sources (de Azevedo et al., 2020). These materials are usually obtained from biopolymers such as starch, chitosan, cellulose and pectin, which can be extracted from agricultural residues or residues generated during food processing (Coelho et al., 2020), helping to reduce their accumulation and provide environmental benefits with a double positive impact (Barone et al., 2021). In addition, these residues are natural sources of bioactive compounds such as fibers, polyphenols, vitamins and minerals (Brito et al., 2020), and can be applied to the production of active packaging, which aim to interact with packaged food, promoting improvements in food security, sensory and nutritional quality (Bhardwaj et al., 2019).

Many researchers already use fruits, vegetables and their residues to produce bioactive films (Dias et al., 2020; Matheus et al., 2020b; Rodríguez et al., 2020; Sá et al., 2020; Melo et al., 2019), however, as far as we know, this research group is the only one of its kind to propose the development of active and biodegradable films based on persimmon (Matheus et al., 2021). Brazil is the fifth largest producer of this fruit in the world and delivers one of the highest yields (Matheus et al., 2020a), thus the use and exploitation of persimmon is encouraged (Sapper et al., 2019; Ramachandraiah et al., 2017) to avoid post-harvest losses during the short and intense harvest (Matheus et al., 2020a; Conesa et al., 2019). Among the value-added products in the context of biorefineries where persmimmon is used as the input, the possibility of obtaining active films is investigated due to the adequate carbohydrate profile and the presence of several active compounds with antioxidant properties such as phenolic compounds, carotenoids and tannins in the fruit (Gea-Botella et al., 2021; Matheus et al., 2020a; Mamet et al., 2017; Zou et al., 2017).

Matheus et al. (2021a and 2021b) were the first to demonstrate the possibility of obtaining flexible films based on persimmon puree with antimicrobial activity against food pathogens, and of using them as part of a minimally processed vegetable package. This study sought to build on these works by adding orange peel flour (OPF) as a bioactive additive and mechanical reinforcement to these films. The analysis focused on the research on antioxidant activity and the profile of volatile compounds, thus demonstrating the study's innovative character. It is noteworthy that during the industrial processing of orange juice, 50% of residue equivalent to the entire original mass is obtained (Bátori et al., 2017) and this vast amount of residue contains a high content of pectin (20 - 30%), dietary fibers, cellulose, hemicellulose, starch, lignin, flavonoids and essential oils (Farahmandfar et al., 2019; Venkatesh and Sutariya, 2019), justifying the incorporation of orange peel into the films.

The objective of this study is to develop antioxidant and aromatic films based on persimmon puree added to orange peel flour and characterize them in terms of their physicochemical, mechanical, optical, antioxidant and volatile component properties.

2. Materials and methods

2.1 Materials

Persimmon fruit (*Diospyros kaki* L.) and orange peel (*Citrus sinensis* 'Pera') were obtained, respectively, from a local agroecological street market and a local market, both in Rio de Janeiro, Brazil. Other reagents and substances used were obtained from commercial sources.

2.2 Preparation of persimmon puree (PP) and orange peel flour (OPF)

Ripe persimmon fruit without injuries were washed, sanitized (200 ppm of sodium hypochlorite, 15 min) and stored at – 12 °C until processing into puree to develop the biodegradable films. This processing consisted of triturating the whole fruit in an industrial blender until homogenous (Matheus et al., 2021).

Orange peels were washed, sanitized, fractionated and dried in a ventilated oven (Marconi, Brazil) at 65 °C for 6 h. After this period, dehydrated orange peels were ground for 5 min in a conventional blender (Philips Walita, Brazil). They were then dried for 1 h at 90 °C and ground again for 1 min. The OPF was packed in aluminized bags and stored at room temperature until further testing (Ferreira et al., 2015).

2.3 Granulometry of OPF

The OPF was characterized as to its granulometry using an electromagnetic sieve shaker (Bertel, Brazil) for 5 min with different meshes (150, 250, 300, 425, 800 and 850 μm), according to Brito et al. (2020) with some adaptations.

2.4 Preparation of biodegradable films

The biodegradable films were prepared by the casting technique. Six film-forming solutions were prepared based on previous studies, using different concentrations of PP, OPF and glycerol as a plasticizer, according to Table 1. All components were mixed until homogenous for three minutes with an industrial blender. Approximately 10 kg m^{-2} of each film-forming solution was dispersed on polystyrene plates and dried in a ventilated oven at 60 ºC for 5 h. The biodegradable films (F1, F2, F3 F4, F5 and F6) were then removed manually from the plates and conditioned at 23 ± 2 °C and 55% of relative humidity in a glass desiccator for 4 d prior to all analyses.

Table 1. Composition of biodegradables films based on persimmon puree (PP) and orange peel flour (OPF) and addition of glycerol (all expressed in g kg^{-1} of film-forming solution).

Film samples	Film-forming solutions		
	PP	**OPF**	**Glycerol**
F1	1000	0	0
F2	980	20	0
F3	920	20	60
F4	900	20	80
F5	900	40	60
F6	880	40	80

2.5 Characterization of biodegradable films

2.5.1 Mechanical properties and thickness

The mechanical properties of the biodegradable films were analyzed using the Universal Testing Machine (EMIC – DL2000, Instron, Brazil) according to the American Society for Testing and Materials method D882-12. The tensile strength (TS), elongation at break (EB) and Young's modulus (YM) were determined, in quintuplicate, for each film sample, using TESC software, version 3.05. The average measurements for each film sample were calculated to obtain the final TS (MPa), EB (%) and YM (MPa) values. Film thickness was measured using a digital micrometer (Digimess, São Paulo, Brazil). The result was given by the average of six different points for each film sample (Andrade et al., 2016).

2.5.2 Water solubility (WS) and water vapor permeability (WVP)

In order to determine the WS of the biodegradable films, the ratio between the initial dry mass (determined by oven drying at 105 °C to constant weight) and final solubilized mass (in distilled water) was obtained. The WVP of the biodegradable films were determined by gravimetric analysis. Both analyses were carried out in triplicate according to Andrade et al. (2016).

2.5.3 Optical properties

Color measurements of the biodegradable films were determined in quintuplicate, using a colorimeter (model CR 400, Konica Minolta Co. Ltd, Japan) to obtain the coordinates L^* (lightness), a^* (+ a^* red to – a^* green) and b^* (+ b^* yellow to – b^* blue), according to Fai et al. (2016). With these results, additional colorimetric parameters were calculated, as follows: total color difference (ΔE^*) (Equation 1) (Nouraddini et al., 2018), and chroma (C^*) (Equation 2) (Fai et al., 2016) is expressed as follows:

$$\Delta E^* = \sqrt{(\Delta L^*)^2 + (\Delta a^*)^2 + (\Delta b^*)^2} \qquad \text{(Eq. 1)}$$

With the following classifications: trace level difference ($0 < \Delta E^* < 0.5$); slight difference ($0.5 < \Delta E^* < 1.5$); noticeable difference ($1.5 < \Delta E^* < 3.0$); appreciable difference ($3.0 < \Delta E^* < 6.0$); large difference ($6.0 < \Delta E^* < 12.0$); and very obvious difference ($\Delta E^* > 12.0$).

$$C^* = \left[\left(a^{*2} + b^{*2}\right)\right]^{1/2} \qquad \text{(Eq. 2)}$$

2.5.4 Scanning electron microscopy (SEM)

The superficial morphology of the biodegradable films was examined by SEM (JSM-6510LV, JEOL, Japan). Film samples were fixed on metal stubs using carbon tape and covered with carbon. The voltage acceleration, magnification and spot size were set at 5 kV, 45 and 1000 x and 15, respectively (Ojagh et al., 2010).

2.5.5 Thermogravimetric analysis

The thermal stability of the biodegradable films was evaluated using TGA (TA Instruments, Q500). The film samples were heated at a constant rate of 20 °C min^{-1}, under nitrogen atmosphere, at a temperature ranging from 50 to 500 °C. The onset temperature was calculated using the Universal Analysis Software 2000, considering the relation between the results at 100 ºC (initial moisture loss) and at the first peak (Matheus et al., 2021).

2.5.6 Fourier transform infrared spectroscopy

FTIR - ATR was performed, in triplicate, using a spectrometer (Perkin-Elmer Spectrum One, USA) equipped with a module of attenuated total reflectance. The sample scanning frequencies were performed at a range of 4000 to 600 cm^{-1}, at a 4 cm^{-1} resolution, collected in 60 scans over the spectra (Matheus et al., 2021).

2.5.7 Antioxidant properties

The obtainment of extracts to analyze the total content of phenolic compounds and the antioxidant activity of the films was based on the work of Rufino et al. (2007). A one gram sample of each film was weighed in a 100 mL beaker. Then, 40 mL of a solution of water:methanol (1:1, v/v) was added to the beaker, homogenized and left to rest for 60 minutes at room temperature. After this period, the supernatant was transferred to a 100 mL volumetric flask and the residue from the first extraction was used in the second extraction cycle, following the same procedure described above, but replacing the aqueous methanol solution with an aqueous acetone solution (70%, v/v). The supernatant from the second extraction was transferred to the same 100 mL volumetric flask, which was completed with distilled water, thus producing the mother solution to carry out the indicated analyses.

2.5.7.1 Determination of total phenolic content (TPC)

The total content of phenolic compounds of the original stock solution described above was evaluated by the Folin-Ciocalteu spectrophotometric method with gallic acid as the reference compound (Hamauzu and Iijima, 1999). The total content of phenolic compounds was calculated from the gallic acid calibration curve (5, 10, 25, 50 and 100 mg L^{-1}) and the results were expressed in mg of gallic acid equivalent (EAG) g^{-1} of the sample. Absorbance was monitored at a wavelength of 760 nm.

2.5.7.2 Determination of antioxidant activity

The determination of the antioxidant activity of the films was carried out using the ABTS reagent: [2,2'-azinobis-(3-ethylbenzothiazoline-6-sulfonate)] (Rufino et al., 2007). The results were expressed as: the concentration of antioxidant that reduces ABTS free radicals by 50%.

2.5.8 Determination of volatile compounds content

2.5.8.1 Sample preparation for chromatographic analysis

Essential oils were extracted by hydrodistillation in a Clevenger apparatus using a 2000 mL flask containing 50g of sample and 1000 mL of distilled water. The isolation process was carried out for four hours at a temperature of 100°C. The essential oil was removed from the Clevenger apparatus by dissolution in ethyl acetate. The residual water present in the extract was eliminated by contact with anhydrous sodium sulfate. The extract was then concentrated with a flow of nitrogen gas to obtain a final volume of 50 µL. This extract was stored in an amber *vial* at a temperature of -18°C until the chromatographic analysis was carried out.

2.5.8.2 Gas chromatography/mass spectrometer system (GC–MS)

The identification of compounds present in the essential oils of orange flour and films F1, F2 and F3 was carried out with the aid of a gas chromatography/mass spectrometry system of the Shimadzu (Japan) type GC-2010Plus/GCMS-QP2010 (Japan) in Busatta et al. (2007). In this process, a fused silica capillary column (30 mx 0.25 mm di.) coated with poly-dimethyl-siloxane (100%) was used, with a film thickness of 0.25 µm (SPB-1, Supelco, USA). The chromatographic oven temperature was initially programmed to rise from 50°C to 100°C at a constant rate of 2°C/minute. Then from 100°C to 145°C at a constant rate of 3°C/minute. Finally, the temperature was increased from 145°C to 280°C at a constant rate of 5°C/minute. Injector, ion source and GC interface temperatures were maintained at 280°C. The mass spectrometer operated at an ionization voltage of 70 eV, scanning the range from 35 to 400 m/z in cycles of three tenths of a second. Helium gas was used as carrier gas at a flow of 1.0 mL/minute. Essential oils diluted in ethyl acetate were injected into the system at a volume corresponding to 1 µL in *Split* of 1:20. The cut-off time for the solvent (ethyl acetate) was established at 3 minutes. The identification of the compounds was based on the comparison of their mass spectra with those available in the NIST12.lib and NIST62.lib libraries, contained in the manager software of this GC/MS system. The identification was complemented by the use of Kovats indices (Van Den Dool and Kratz, 1963). These indices were estimated with the aid of a mixture of saturated alkanes (C_9-C_{26} – 1.000 µg of each component / mL of hexane).

2.5.8.3 Gas chromatography with a flame ionization detector (GC-FID)

The volatile compounds contained in essential oils diluted in ethyl acetate were separated and quantified using a gas chromatography system containing a flame ionization detector (GC-2010Plus). Column and chromatographic conditions were the same as described above. The detector temperature was set at 280°C. The quantification of volatile compounds in these matrices was conducted using the area normalization technique.

2.6 Statistical Analyses

Statistical analyses were performed using the Graph Pad Prism 7.0 software. The data obtained underwent a simple analysis of variance (one-way ANOVA), with Tukey's post-test for multiple comparisons between groups. The Pearson correlation test was also used to assess the correlation between different variables. A level of $p < 0.05$ was considered statistically significant.

3. Results and discussion

3.1 Characterization of OPF fractions

The OPF fractions of 800-850 μm, 250-425 μm and < 150μm represented 2.7%, 39.09% and 56.21% of the total flour, respectively. The OPF fractionation showed that the highest percentage of flour remained in the finest fraction (<150μm). Thus, the flour fraction <150 μm was used for addition in biodegradable films, as it represents the highest percentage of flour retention and because we believe that using the finest flour fraction would result in better dispersion in the polymer matrix. Furthermore, studies show that the incorporation of food residues in the form of flour in biodegradable films, to the detriment of the extract, provides the films with better mechanical resistance (Luchese et al., 2021).

3.2 Mechanical properties

The corresponding films were continuous, homogeneous, manageable and orange in color are shown in Figure 1. Table 2 shows the results on barrier, mechanical and optical properties. Similar results to this study were found in other fruit-based films in the literature (Matheus et al., 2021; Neto et al., 2018; Barros-alexandrino et al., 2018; Jirukkakul, 2016). Some studies show that the incorporation of fibers in biodegradable polymers improves TS values (Kevij et al., 2020; Espitia et al., 2014), as observed in F2. F2 is the film that presents in its composition PP and the addition of OPF (source of pectin according to Han et al. (2021)) without presence of plasticizer (glycerol), and this formulation resulted in a more compact and cohesive matrix. The improvement in TS values associated with pectin is due to its high chemical affinity with fruit puree (Viana et al., 2018). In addition, its presence in cellulosic media provides firmness and mechanical resistance to the polymer matrix due to the intermolecular bonds between homogalacturonic regions and cellulosic microfibrils (Martelli et al., 2014).

The glycerol content has complex effects on the mechanical properties of the films. This compound interacts with the polymer matrix, breaking the hydrogen bonds that provide stability to the chain (Nogueira et al., 2018). Such interactions generate free volumes between the chains, decreasing their intermolecular forces, causing the structural disruption of the matrix (plasticizing action) and favoring molecular mobility (Martelli et al., 2013). There is a significant difference between the F1 and F4 films, due to the fact that the F4 film has both a lower proportion of OPF and a higher glycerol content, which provides, considering all other films, higher EB values, preserving flexibility of the films. The EB found in this study is in the same range as in other studies, namely 13.15% for papaya puree (Tulamandi et al., 2016); 13% for banana puree (Martelli et al., 2013); 10% for cupuaçu puree (Melo et al., 2019); 14.99% for fruit and vegetable residues (Hafeez et al., 2021); and 10.77% for apple pomace (Gustafsson et al., 2019).

Films obtained from fruit puree tend to have lower YM values due to the matrix dilution caused by the fruit components, which plays a similar plasticizer role to that of glycerol (Viana et al., 2018), as observed in the F3 (4.51 MPa), F4 (3.36 MPa), F5 (3.48 MPa) and F6 (1.8 MPa) films. Other studies found lower stiffness values for fruit puree films, such as 3.82 MPa for persimmon films (Matheus et al., 2021), and 4.13 MPa for banana peel flour and corn starch films (Arquelau et al., 2019).

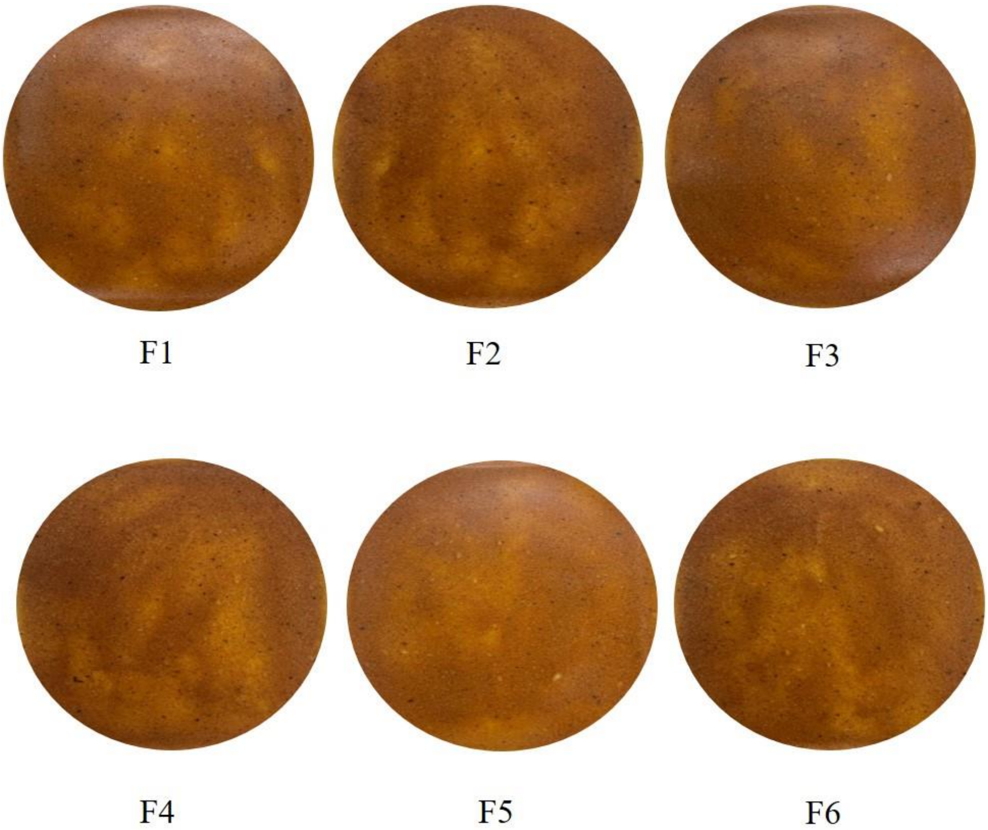

Figure 1. Visual aspects of the films: F1 (formulated using 1000 g kg^{-1} persimmon); F2 (formulated using 980 g kg^{-1} persimmon, 20 g kg^{-1} of OPF); F3 (formulated using 920 g kg^{-1} persimmon, 20 g kg^{-1} of OPF and 60 g kg^{-1} glycerol); F4 (formulated using 900 g kg^{-1} persimmon, 20 g kg^{-1} of OPF and 80 g kg^{-1} glycerol); F5 (formulated using 900 g kg^{-1} persimmon, 40 g kg^{-1} of OPF and 60 g kg^{-1} glycerol); and F6 (formulated using 880 g kg^{-1} persimmon, 40 g kg^{-1} of OPF and 80 g kg^{-1} glycerol).

3.3 WS and WVP

The WS of the films ranged from 45.27% to 75.8%. Films F3 and F4 showed a tendency to high WS values, which may be related to a higher glycerol / FPO ratio, while F5 showed an inverse tendency with lower WS values, which can be attributed to its greater thickness. The increased solubility of polysaccharide-based films is attributed to the hydrophilic nature of glycerol and other polar compounds present in the polymer matrix (Singh et al., 2018). These molecules interact with water, facilitating transport and increasing solubility (Arquelau et al., 2019). The ideal value for film solubility depends on its application or proposed use (Singh et al., 2018). When the WS of a film is high, its ability to protect food from a high humidity environment is diminished; however, it is advantageous from the point of view of its biodegradability (Capitani et al., 2016). Other studies showed similar results for films with fruit extract: 65.3% for acerola, 73.1% for cashew and 70.5% for strawberry (Eça et al., 2015); and 45.24 to 52.28% for pineapple bagasse films (Susmitha et al., 2021).

The presence of hydrophilic components such as proteins, carbohydrates and fibers in the polymer matrix leads to greater spacing between the molecules, which facilitates the diffusion of water through the film, with a consequent increase in permeability (Silva et al., 2020). According to Otoni et al. (2014), it is observed that the addition of plasticizers contributes to increased permeability by weakening the intermolecular interactions between polymer chains. In addition to the material's hydrophobicity, film thickness also influences WVP. Thinner hydrophilic films are less permeable due to the lower affinity to water compared to thicker films (Stoll et al., 2017). The films in, general, had high WVP values, without expressive difference between them. Other studies showed similar WVP values: 3.5×10^{-6}, 6.64×10^{-6}, and 8.31×10^{-6} g h^{-1} m^{-1} Pa^{-1} for papaya puree films (Barros-Alexandrinho et al., 2019), persimmon puree (Matheus et al., 2021) and mango puree with nanofibrillated bacterial cellulose (Viana et al., 2018), respectively.

Table 2. Film characterization: barrier, mechanical, optical, antioxidant properties, and total phenolic content.

Barrier and mechanical properties	**F1**	**F2**	**F3**	**F4**	**F5**	**F6**
Water vapor permeability (x 10^{-6} g h^{-1} m^{-1} Pa^{-1})	7.72±1.41^{b,c}	7.09±0.12^{c}	10.75±1.65^{a}	10.82±0.17^{a}	10.39±0.74^{a,b}	12.61±1.30^{a}
Water solubility (%)	62.02±0.29^{a,b}	55.23±7.73^{a,b}	72.25±7.95^{a}	75.80±11.45^{a}	45.27±10.12^{b}	65.99±8.67^{a,b}
Thickness (mm)	0.59±0.01^{d}	0.74±0.01^{b}	0.65±0.03^{c}	0.74±0.02^{b}	0.82±0.01^{a}	0.72±0.02^{b}
Tensile strength (MPa)	0.74±0.19^{b}	1.71±0.37^{a}	0.66±0.09$^{b, c}$	0.4±0.09$^{b, d}$	0.34±0.12$^{c, d}$	0.2±0.06^{d}
Elongation at break (%)	10.24±0.79^{b}	10.5±1.54$^{a, b}$	15.32±2.99$^{a, b}$	15.84±4.77^{a}	12.69±2.1$^{a, b}$	13.74±2.6$^{a, b}$
Young's modulus (MPa)	11.93±0.96^{b}	15.32±0.98^{a}	4.51±1.71^{c}	3.36±0.2$^{c, d}$	3.48±0.52$^{c, d}$	1.8±0.62^{d}
Optical properties	**F1**	**F2**	**F3**	**F4**	**F5**	**F6**
*L**	61.38±1.80^{a}	53.26±0.89^{c}	53.90±1.13$^{b, c}$	55.87±0.81^{b}	53.12±1.29^{c}	53.72±0.92$^{b, c}$
*a**	18.95±0.83^{a}	15.08±0.93^{c}	17.03±0.40^{b}	18.73±0.49^{a}	14.89±0.62^{c}	16.00±1.08$^{b, c}$
*b**	47.79±2.00^{a}	35.29±1.26^{d}	38.50±1.54^{c}	42.28±1.40^{b}	35.88±1.45$^{c, d}$	37.48±1.58$^{c, d}$
ΔE*	61.2±1.05^{a}	57.21±0.87^{c}	59.19±0.57^{b}	60.71±0.65$^{a, b}$	57.62±0.73^{c}	58.42±0.72$^{b, c}$
Chroma	51.42±1.71^{a}	38.38±1.51^{d}	42.10±1.47^{c}	46.24±1.45^{b}	38.85±1.56^{d}	40.76±1.83^{c,d}
Antioxidant activity	**F1**	**F2**	**F3**	**F4**	**F5**	**F6**
TPC (mg EAG/g)	4.0±0.2^{c,d}	5.3±0.2^{a}	3.8±0.1^{d}	3.8±0.1^{d}	4.6±0.0^{b}	4.4±0.1^{b,c}
$IC_{50(ABTS)}$ (µg/mL)	1818.2±189.1^{c}	919.9±40.5^{d}	2410.4±105.4^{a,b,c}	2747.0±55.2^{a}	2571.6±31.6^{a,b}	1972.8±347.7^{b,c}

PP and OPF biodegradable films: F1 (formulated using 1000 g kg^{-1} persimmon); F2 (formulated using 980 g kg^{-1} persimmon, 20 g kg^{-1} of OPF); F3 (formulated using 920 g kg^{-1} persimmon, 20 g kg^{-1} of OPF and 60 g kg^{-1} glycerol); F4 (formulated using 900 g kg^{-1} persimmon, 20 g kg^{-1} of OPF and 80 g kg^{-1} glycerol); F5 (formulated using 900 g kg^{-1} persimmon, 40 g kg^{-1} of OPF and 60 g kg^{-1} glycerol); and F6 (formulated using 880 g kg^{-1} persimmon, 40 g kg^{-1} of OPF and 80 g kg^{-1} glycerol).

Results expressed as mean ± standard deviation of three replicates. Means followed by the same letter within a column indicate no significant ($p < 0.05$) difference among samples.

3.4 Optical properties

The color of packaging material plays an important role in consumer acceptance of food products. Table 2 presents the values of *a** and *b**, indicating that the films are largely red and yellow, which demonstrates a natural tendency to maintain the color of the persimmon pulp, as seen in Figure 1. Furthermore, these values reflect the presence of carotenoids and flavonoids, responsible for the yellow and orange coloration, present in the orange peel (Deng et al., 2019). Film brightness is defined by *L** values, which range from 0 (black color) to 100 (white color). F1 had a higher *L** value, which may be related to the absence of pectin and glycerol in the filmogenic solution, allowing more light to pass through the film and consequently increasing the clarity of the films (Fakhouri et al., 2015). In addition, the thickness influenced the luminosity. Thinner films tended to increase in *L** (F1). Films F2, F5 and F6 had lower values of *C**, which reflects the film's color saturation, ranging from opaque (low value) to vivid (high value) (Fai et al., 2016). According to Kevij et al. (2020), the increase in the opacity of these films is directly related to the increase in the concentration of OPF in the polymer matrix.

3.5 SEM

The morphological aspects of the surface of the films were analyzed by SEM and the micrographs are shown in Figure 2. Biodegradable films have a microstructure with compact and homogeneous surfaces. The high concentration of pectin forms an extensive intermolecular compaction network, which can cause visible cracks on the film’s surface (Sucheta et al., 2019), but was not observed in this study. As glycerol was added, the surfaces became rougher; a rougher matrix evidences the behavior of the plastic (Matheus et al., 2021; Martelli et al., 2013). No plasticizer agglomerate was identified in the films, indicating a uniform dispersion in the puree matrix.

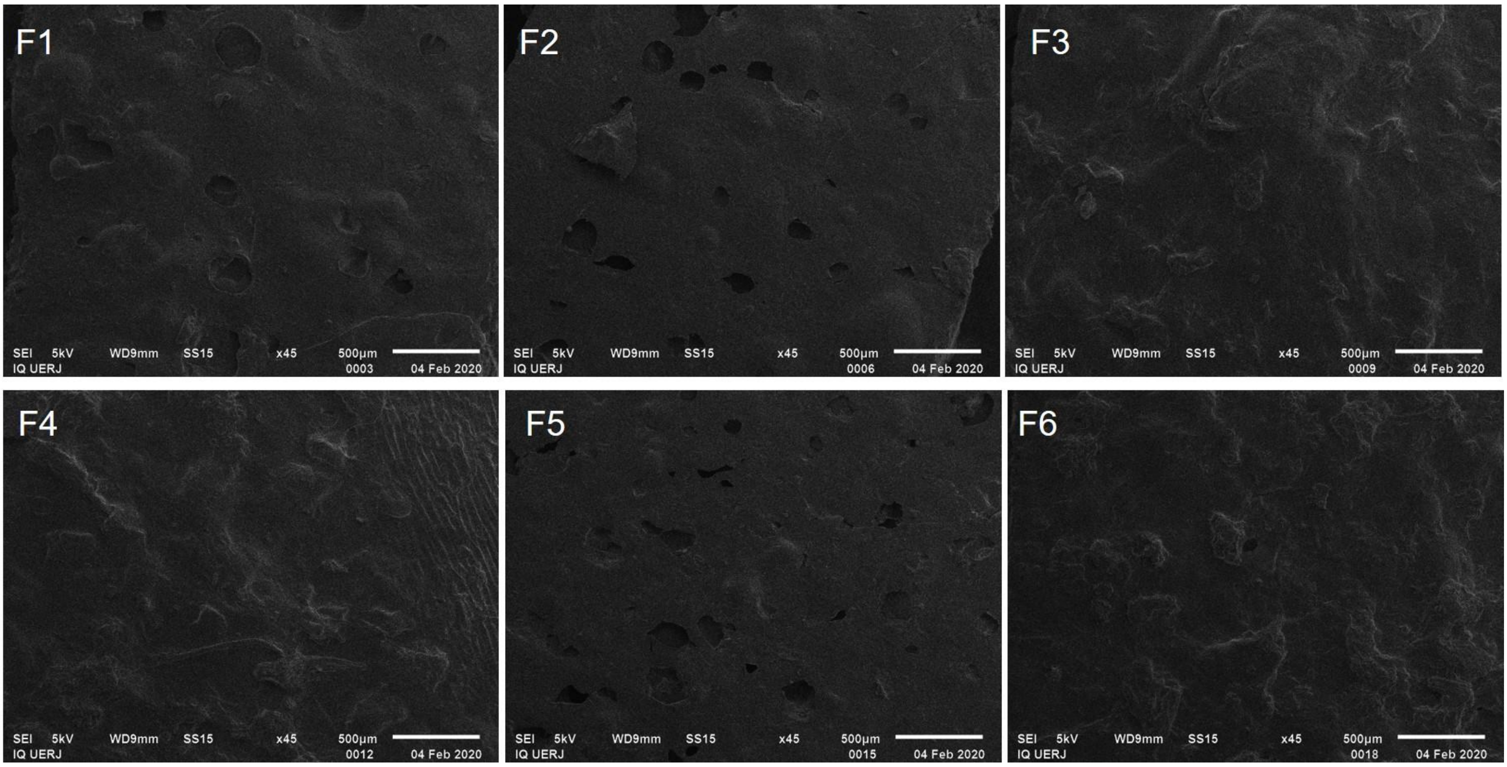

Figure 2. Scanning electron microscopy images of the surface of biodegradable films using a x45 magnitude. F1 (formulated using 1000 g kg^{-1} persimmon); F2 (formulated using 980 g kg^{-1} persimmon, 20 g kg^{-1} of OPF); F3 (formulated using 920 g kg^{-1} persimmon, 20 g kg^{-1} of OPF and 60 g kg^{-1} glycerol); F4 (formulated using 900 g kg^{-1} persimmon, 20 g kg^{-1} of OPF and 80 g kg^{-1} glycerol); F5 (formulated using 900 g kg^{-1} persimmon, 40 g kg^{-1} of OPF and 60 g kg^{-1} glycerol); and F6 (formulated using 880 g kg^{-1} persimmon, 40 g kg^{-1} of OPF and 80 g kg^{-1} glycerol).

3.6 TGA

The thermal stability of biodegradable films was investigated by TGA. All samples show two important stages of thermal degradation during the analysis, which varied between 210°C and 340°C (Figure 3). Thermograms show the gradual loss of mass at the beginning of heating, at around 100°C, which is due to the evaporation of free water, which is naturally retained in the matrix of the films, and to the low molecular weight volatile compounds (Xu et al., 2017). Films that contain glycerol in their composition have a higher percentage of mass loss between 171°C and 243°C, evidencing the thermal degradation of glycerol in this temperature range (130°C - 230°C), in addition to the decomposition of pectin at 226°C (Mendes et al., 2019; Espitia et al., 2014). The point above 300°C is very similar for all samples and is related to decomposition of the polymer structure and organic compounds, through the degradation of the monosaccharide rings (Wang et al., 2018; Martelli et al., 2013).

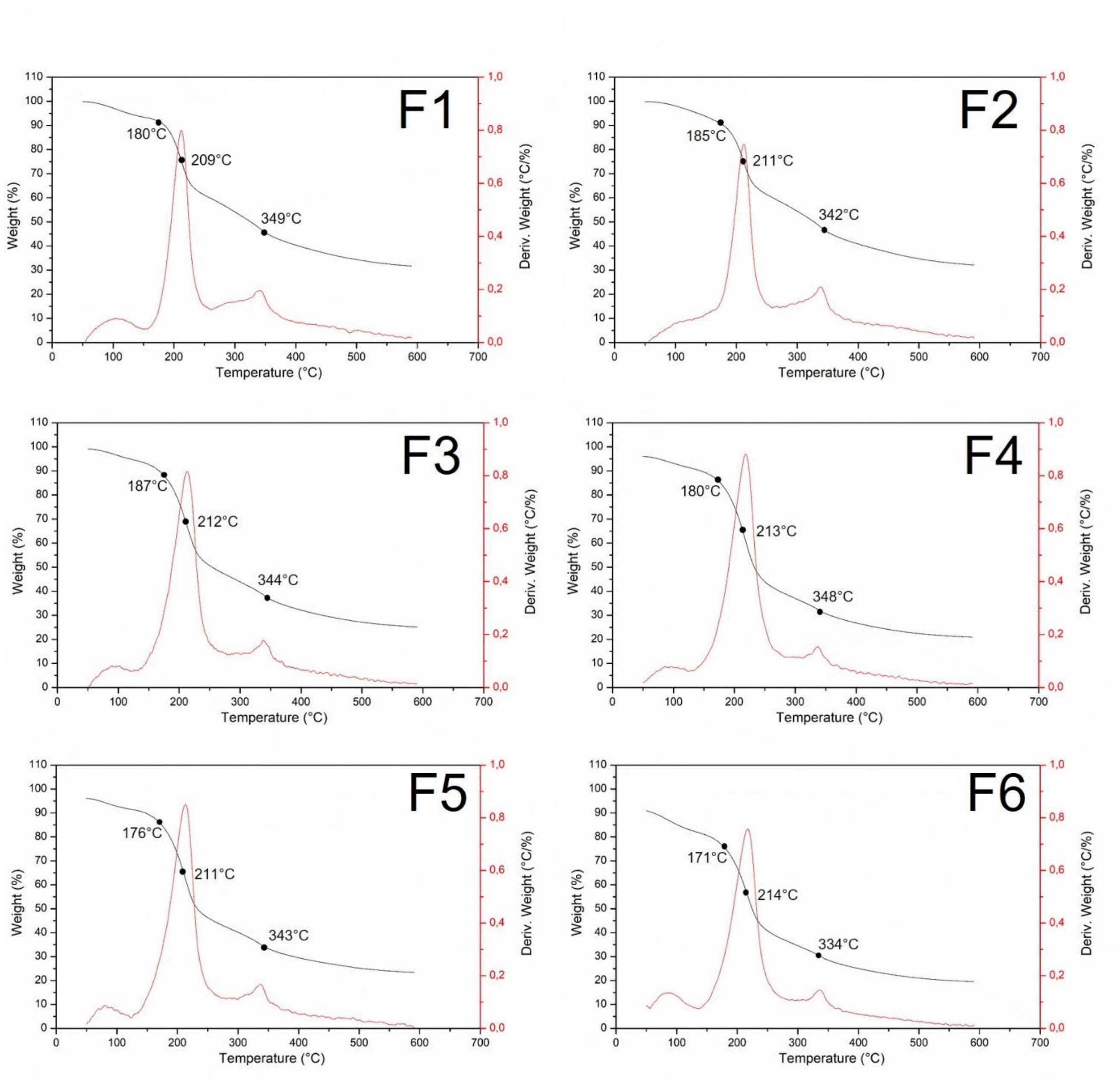

Figure 3. The TGA curve of PP and OPF films: F1 (formulated using 1000 g kg^{-1} persimmon); F2 (formulated using 980 g kg^{-1} persimmon, 20 g kg^{-1} of OPF); F3 (formulated using 920 g kg^{-1} persimmon, 20 g kg^{-1} of OPF and 60 g kg^{-1} glycerol); F4 (formulated using 900 g kg^{-1} persimmon, 20 g kg^{-1} of OPF and 80 g kg^{-1} glycerol); F5 (formulated using 900 g kg^{-1} persimmon, 40 g kg^{-1} of OPF and 60 g kg^{-1} glycerol); and F6 (formulated using 880 g kg^{-1} persimmon, 40 g kg^{-1} of OPF and 80 g kg^{-1} glycerol).

3.7 FTIR

FTIR analysis showed that the sample spectra were similar in all absorption peaks associated with pectin structures such as OH elongation (3300 cm^{-1}) and C - O elongation (1102 cm^{-1}) and saccharide bonds (1165 cm^{-1}) (Basiak et al., 2018; Oliveira et al., 2017). Small differences were noted and are represented in Figure 4. Absorption bands close to 1620 cm^{-1} are attributed to the elongation of pectin free carboxylic groups (C=O) (Chodijah et al., 2019), while 1630 cm^{-1} and 1650 cm^{-1} are related to the individual components of the biopolymer and glycerol, respectively (Liu et al., 2013). It is suggested that the addition of glycerol to biodegradable films promotes interactions of hydrogen bonds with pectin due to the observed changes in magnitude; this was also described by Liu et al. (2013) regarding starch, chitosan and glycerol films, where there were changes in peak absorption positions when glycerol was added.

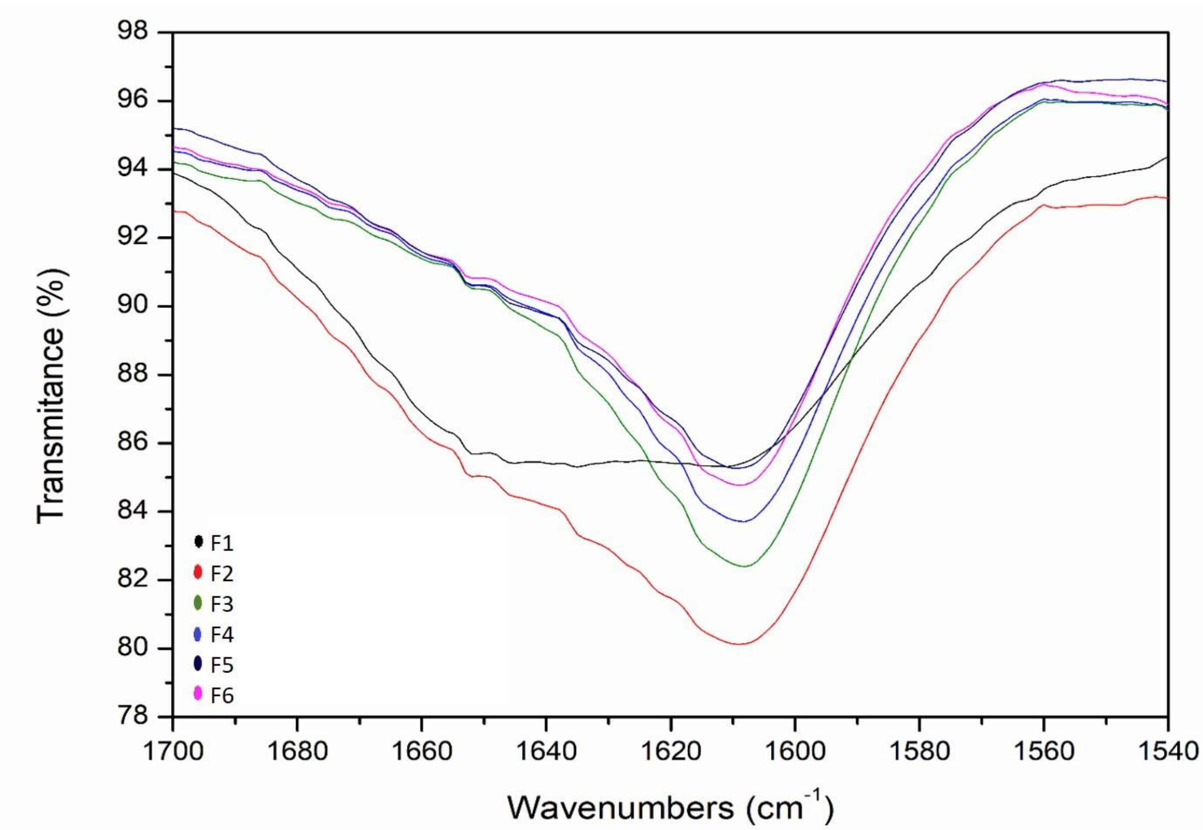

Figure 4. FTIR off PP and OPF films: F1 (formulated using 1000 g kg^{-1} persimmon); F2 (formulated using 980 g kg^{-1} persimmon, 20 g kg^{-1} of OPF); F3 (formulated using 920 g kg^{-1} persimmon, 20 g kg^{-1} of OPF and 60 g kg^{-1} glycerol); F4 (formulated using 900 g kg^{-1} persimmon, 20 g kg^{-1} of OPF and 80 g kg^{-1} glycerol); F5 (formulated using 900 g kg^{-1} persimmon, 40 g kg^{-1} of OPF and 60 g kg^{-1} glycerol); and F6 (formulated using 880 g kg^{-1} persimmon, 40 g kg^{-1} of OPF and 80 g kg^{-1} glycerol).

3.8 Antioxidant properties

TPC analysis is used to assess the amount of phenolic compounds present in film samples. Phenolic content affects the antioxidant activity which prevents oxidation (Yuan et al., 2015), therefore the TPC of PP and PP/OPF films were examined (Table 2). The TPC of PP films increased 32.5% and 15% according to the addition of higher concentrations of OPF in the filmogenic solution, F2 and F5 respectively, most likely due to the presence of a variety of polyphenols such as anthocyanins, flavonoids, tannins and procyanidins (Jridi et al., 2019). Among the most abundant flavonoids, hesperidin, tangeretin and naringenin, typical of the citrus fruit peel, stand out, in addition to phenolic acids such as ferulic, coumaric and synapic acid (Jridi et al., 2019; Barrales et al., 2018). The same was observed by Kevij et al. (2020) in gelatin films incorporated with different concentrations of orange peel powder, where there was an increase in TPC proportional to the addition of orange peel. However, it is observed that F2 had higher TPC although the concentration of OPF was lower compared to F5 and F6. This evidences the action of glycerol in the polymeric matrix, with the assumption that the matrix components become more diluted and more dispersed with the addition of a plasticizer, resulting in a lower quantifiable content of phenolics. These results are in accordance with those of Adilah et al. (2018) who stated that the TPC of gelatin films improved after the incorporation of mango peel extract.

Due to the different mechanisms of antioxidant activity and varieties of bioactive compounds present in natural residues, other antioxidant assays were performed in this study. The F1 film (100% PP) showed antioxidant activity due to the presence of tannins, carotenoids and phenolic compounds inherent in the composition of persimmon (Matheus et al., 2020), however, the incorporation of OPF in the PP film improved the antioxidant activity of the films. After the addition of 2% OPF in the PP (F2) films there was an increase in the ABTS radical scavenging capacity of the films compared to F1 (100% PP), the same was not observed when glycerol was added to the polymer matrix, even at concentrations of 4% of OPF (F5 and F6). Phenolic compounds are associated with metal ion chelation and radical scavenging activities, which confers their antioxidant abilities (Lorenzo et al., 2018). Furthermore, the antioxidant activity of the PP films, which increased with the addition of OPF, corresponded to the increase in the TPC of the films, confirmed by Pearson's Correlation Test ($r = -0.74$ and $p<0.05$). Han and Song (2021) obtained similar results for watermelon peel pectin (WRP) films containing kiwi peel extract (KPE), where the incorporation of KPE increased the ABTS radical scavenging capacity of the films, which was associated with active compounds present in natural extracts. Hanani et al. (2018) observed the same trend for fish gelatin films added to pomegranate peel powder (PPP). The incorporation of 5% of PPP in the films increased ABTS radical scavenging activity by 80%, suggesting that the phenolic compounds present in PPP are capable of scavenging free radicals and metal chelate cations.

3.9 Volatile composition

Given the results presented above, the F1, F2, and F3 films were chosen to analyze the profile of volatile compounds since they offered the most promising results in terms of mechanical and barrier properties, total phenolic content, and antioxidant activity.

The yields of the isolation processes of volatile compounds (essential oils) from orange peel flour (OPF) and F1, F2 and F3 films are shown in Table 3. The results are expressed in g of essential oil per 100 g of sample. In general, the yields were extremely low, with the OPF essential oil yield being higher than the others ($p < 0.05$).

Table 3. Yields of the extraction processes of essential oils from OPF and F1, F2 and F3 films.

Samples	Yield (g of essential oil/100 g of sample)
OPF	0.0693 ± 0.0024[a]
F1	0.0419 ± 0.0073[b]
F2	0.0270 ± 0.0042[b]
F3	0.0245 ±0.0064[b]

Results expressed as mean ± standard deviation of three replicates. Mean values followed by the same letter within a column indicate no significant ($p < 0.05$) difference among samples.

Table 4 shows the essential oil profiles of the analyzed samples. A total of 71 substances were identified as components of one or more of these samples. The group of non-terpenic compounds contains the greatest number of representatives, with 39 components. The group of terpenic compounds has 32 members. The first group consists of 10 esters, 2 alcohols, 17 carboxylic acids, 6 aldehydes, 1 aromatic compound, 1 hydrocarbon, 1 ketone and 1 carboxylic acid derivative. The group of terpene compounds is formed by 2 monoterpenes, 7 oxygenated terpenes, 7 sesquiterpenes and 16 oxygenated sesquiterpenes. OPF essential oils have 69 compounds, representing 97.18% of the total of 71 compounds identified in this study. The essential oil isolated from F1 displays the poorest chemical profile; only 25 volatile compounds were detected in this essential oil. The essential oil from F2 and F3, in turn, presented 60 and 51 components in their constitution, respectively. Carpes et al. (2021) also identified a range of volatile compounds (56) in cassava starch films added to apple pomace (AP), including 13 esters, 13 aldehydes, 9 ketones and 7 alcohols. This study showed that AP was responsible for 75% of the volatile compounds identified, reinforcing that the incorporation of food residues in bio-based films contributes to the improvement of their aromatic and bioactive properties.

By analyzing the results mentioned above, it is possible to infer that OPF is the main source of volatile compounds isolated from F2 and F3 films. This flour is responsible for the presence of most terpenic compounds found in these two films. Several of these compounds have the potential to influence the overall aroma of these matrices, as they have low odor thresholds. The odor threshold value is an important characteristic to estimate the sensory relevance of a compound in a medium. This parameter is defined as the minimum concentration of the volatile substance that can be perceived by human olfaction. The lower the odor threshold value of a substance, the greater its odor potential.

Limonene, for example, the main terpene compound found in OPF and in F2 and F3 films, has an odor threshold in water of 10 $\mu g\ L^{-1}$ and a fresh aroma associated with that emanating from pine trees (Teixeira et al., 2020). Thus, due to its low odor threshold value and its relatively high concentration in these matrices, this compound has great potential to positively influence the aroma of these products. α-Sinensal, with its woody, green and grassy aroma, has a very low odor threshold (0.05 $\mu g\ L^{-1}$) and, therefore, even present in low concentrations, it can also influence the aroma of OPF and F2 and F3 films (Sawamura et al., 2004; Ohloff, 1978). β-Myrcene, with an odor threshold in water ranging from 13 – 15 $\mu g\ L^{-1}$ and an aroma characterized as spicy, herbaceous and citrus (Teixeira et al., 2020) also has the potential to interfere with the aroma of these three products (OPF, F2 and F3). Likewise, it is possible to infer that the sesquiterpene known as caryophyllene (odor threshold in water of 64 μg L^{-1}), with a woody-spiced, dry and greasy aroma may also contribute to the aroma of these matrices (Teixeira et al., 2020). Nerolidol and β-linalool, detected in OPF and F2 film, may also play an important role in the aroma of these products. Nerolidol is associated with a woody, apple, sweet and citrus odorous notes and its odor threshold in water is 12 μg L^{-1}. β-Linalool has orange, citrus and floral aroma and an odor threshold in water of 6 μg L^{-1} (Teixeira et al., 2020).

β-ionone is another terpene compound that deserves to be highlighted due to its very low odor threshold value in water (0.007 μg L^{-1}). The aroma of this substance can be described as fruity, with floral, perfume and soapy notes (Brito et al., 2021). This active odor oxygenated monoterpene was detected in all analyzed samples. Thus, its origin is associated with both OPF and persimmon. Geranyl acetone and farnesyl acetone were also detected in all samples (OPF, F1, F2 and F3), so the presence of both in the films must also be associated with both OPF and persimmon. Geranyl acetone (oxygenated monoterpene) has a sweet, woody, persimmon aroma and an odor threshold in water of 60 μg L^{-1} (Wang et al., 2011; Buttery et al., 1988). Unfortunately, it was not possible to find any information in the scientific literature on the odor threshold and sensory characteristics of farnesyl acetone (oxygenated sesquiterpene). Geranyl acetone, β-ionone and farnesyl acetone were the only terpenic compounds found in the F1 film, which is composed of 100% persimmon. These three terpenic compounds are degradation products of carotenoids. β-Ionone appears to originate from the oxidative breakdown of β-carotene, while farnesyl acetone and geranyl acetone are probably derived from phytoene or phytofluene (Lewinsohn et al., 2005). Geranyl acetone and β-ionone have previously been detected in persimmon samples (Wang et al., 2011).

Some non-terpenic compounds can also be classified as active odor substances capable of influencing the overall aroma of the products evaluated in this study. This is the case, for example, of nonanal (OPF and F2), dodecanal (OPF, F2 and F3) and isovaleric acid (OPF, F2 and F3). Nonanal (1 μg L^{-1} of odor threshold in water) has a fresh, citrusy odor, while dodecanal is associated with a green, grassy aroma and has an odor threshold in water of 2 μg L^{-1}. Isovaleric acid has a cheese-like aroma and an odor threshold in water of 120 μg L^{-1} (Ohloff, 1978; Boelens and Gemert, 1987; Buttery et al., 1988; Sawamura et al., 2004).

In addition to the influence on the aroma of the films, several of the compounds identified as components of the volatile fraction of these matrices are capable of providing interesting bioactive properties for this type of coating material. In this sense, the terpenic compounds mainly originating from the OPF stand out. Nerolidol, for example, detected in OPF and in the F2 film, has antioxidant activity and antimicrobial action (Teixeira et al., 2020). α-Terpineol, which was detected in OPF and in F2 and F3 films, also has antimicrobial potential, as does limonene (OPF, F2 and F3). Furthermore, this monoterpene (limonene) boasts antiviral activity (Teixeira et al., 2020; Mariano et al., 2019). Caryophyllene (OPF, F2 and F3) and terpinen-4-ol (OPF and F2) show bactericidal activity and β-linalool (OPF and F2) and germacrene D (F2) show bactericidal and fungicidal actions (Teixeira et al., 2020; Mariano et al., 2019). These compounds can exert their bioactive potential independently or synergistically. The properties (antioxidant, bactericide, fungicide), attributed to some of these compounds, have the potential to increase the shelf life of food products when covered with these films.

The addition of glycerol in the production of the F3 film caused a dilution of the persimmon puree and the OPF used in the filmogenic solution. This caused the non-detection of some compounds that had been identified in the OPF and in the F2 film. Although the addition of glycerol confers benefits on mechanical properties, this dilution can influence the aroma of this matrix and also its bioactive properties.

Despite the importance of the terpenic compounds mentioned above, the quantitative evaluation of the essential oils of these products highlighted the dominant compounds as the carboxylic acids and esters. For example, the major compounds of OPF were palmitic acid [(40.71 ± 0.45)%], linolenic acid [(27.01 ± 0.13)%], methyl linoleate [(4.12 ± 0.04)%] and methyl palmitate [(3.29 ± 0.03)%]. In the case of the films (F1, F2 and F3) the esters were replaced in quantitative importance by vaccenic acid and tetradecanoic acid (Table 4). Finally, the presence of the essential fatty acid linolenic acid in the OPF and in the three films analyzed is noteworthy, highlighting the presence of bioactive substances with high added value in these products.

Table 4. Volatile compounds identified in essential oils of OPF and F1, F2, and F3 films.

№	Compounds	RT	IKc	IKl	OPF (%)	F1 (%)	F2 (%)	F3 (%)
1	Isobutyl acetate[Es]	3.35	---	---	0.20±0.01	0.025±0	0.27±0.01	0.24±0.01
2	3-methyl-2-Buten-1-ol[A]	3.40	---	---	0.18±0	---	---	0.005±0
3	Valeric acid[Ac]	3.75	---	---	0.005±0	0.005±0	0.01±0	0.01±0
4	Furfural[Al]	4.21	---	---	0.21±0.01	---	0.005±0	0.01±0
5	Isovaleric acid[Ac]	5.14	---	---	0.01±0	---	0.02±0	0.02±0
6	Ethylbenzene[CA]	5.40	---	---	0.03±0	---	0.03±0	0.025±0
7	o-Xylene[H]	5.67	---	---	0.02±0	---	0.03±0	0.02±0
8	5-Methyl-2-furfural[Al]	8.85	930	932[NIST]	0.06±0	---	---	---
9	beta-Myrcene[M]	11.06	975	975[NIST]	0.02±0	---	0.01±0	0.005±0
10	D-Limonene[M]	13.07	1013	1013[NIST]	1.97±0.07	---	1.2±0.07	0.67±0.04
11	Nonanal[Al]	16.92	1076	1076[NIST]	0.01±0	---	0.06±0	0.005±0
12	beta-Linalool[MO]	17.09	1078	1078[NIST]	0.19±0	---	0.03±0.01	---
13	Limonene oxide[MO]	18.04	1094	1106[NIST]	0.09±0.01	---	0.02±0	---
14	L-Terpinen-4-ol[MO]	21.71	1148	1148[NIST]	0.05±0	---	0.01±0	---
15	alpha-Terpineol[MO]	22.54	1161	1161[NIST]	0.24±0	---	0.04±0	0.01±0
16	cis-Carveol[MO]	24.46	1189	1188[NIST]	0.63±0.01	---	0.04±0	---
17	1-Decanol[A]	28.63	1255	1255[NIST]	0.26±0.02	---	0.035±0.01	---
18	Copaene[SO]	34.43	1358	1358[NIST]	0.1±0	---	0.12±0	0.03±0
19	n-Decanoic acid[Ac]	35.06	1370	1370[NIST]	0.42±0.08	---	0.06±0	---
20	Dodecanal[Al]	35.61	1381	1383[NIST]	0.27±0.01	---	0.08±0	0.05±0.01
21	Caryophyllene[S]	36.39	1395	1395[NIST]	0.15±0	---	0.05±0	0.01±0
22	beta-Cubebene[S]	36.90	1406	1390[Pherobase]	0.10±0	---	0.03±0	0.03±0
23	cis-Geranylacetone[MO]	37.53	1419	1424[NIST]	0.02±0	0.01±0	0.03±0	0.01±0
24	(2-Hexylcyclopropyl)acetic acid[Ac]	38.81	1447	1435[ChemSpider]	0.10±0.05	---	0.06±0	0.02±0
25	trans-beta-Ionone[MO]	38.92	1449	1452[NIST]	0.06±0.01	0.02±0	0.05±0	0.02±0
26	Undecanoic acid[Ac]	39.64	1465	1465[NIST]	0.14±0.04	---	0.005±0	---

27	Eremophilene[S]	39.85	1469	1486[Pherobase]	0.28±0.02	---	0.12±0	0.02±0
28	Ledene[S]	39.93	1471	1489[Pherobase]	0.04±0	---	0.01±0	---
29	alpha-Muurolene[S]	40.20	1477	1478[NIST]	0.06±0	---	0.03±0	0.01±0
30	Germacrene D[S]	40.66	1487	1487[NIST]	---	---	0.02±0	---
31	delta-Cadinene[S]	41.17	1498	1509[NIST]	0.68±0	---	0.21±0	0.09±0.01
32	Elemol[SO]	41.97	1519	1529[PubChem]	0.27±0	---	0.05±0	0.01±0
33	trans-Nerolidol[SO]	42.71	1539	1539[Pherobase]	0.35±0.01	---	0.1±0	---
34	Spathulenol[SO]	42.93	1545	1575[Pherobase]	0.04±0	---	---	---
35	Caryophyllene oxide[SO]	43.07	1549	1549[NIST]	0.23±0	---	---	---
36	Dodecanoic acid[Ac]	43.60	1564	1568[Pherobase]	1.34±0	0.04±0	0.30±0	0.18±0.01
37	Globulol[SO]	44.67	1593	1593[NIST]	0.37±0.06	---	0.07±0.01	0.04±0
38	Cubenol[SO]	44.76	1595	1595[NIST]	0.05±0.01	---	0.03±0.01	0.01±0
39	gama-Eudesmol[SO]	44.87	1598	1598[PubChem]	0.28±0	---	0.06±0.01	0.025±0
40	tau-Cadinol[SO]	45.23	1610	1610[NIST]	0.06±0.01	---	0.04±0	0.01±0
41	delta-Cedrol[SO]	45.32	1613	1614[NIST]	0.23±0	---	0.06±0.01	0.025±0
42	beta-Eudesmol[SO]	45.42	1616	1630[Pherobase]	0.36±0.02	---	0.11±0.02	0.02±0
43	alpha-Eudesmol[SO]	45.59	1622	1649[Pherobase]	0.6±0	---	0.15±0.01	0.03±0
44	Tridecanoic acid[Ac]	46.40	1649	1648[NIST]	0.13±0	---	---	---
45	2,6,10-Trimethyl-2,6,9,11-dodecatetraenal[Al]	46.76	1661	1686[Pherobase]	0.23±0	---	---	---
46	n-Pentadecanal[Al]	47.47	1684	1687[NIST]	0.02±0.02	0.02±0	0.15±0	0.04±0.02
47	trans-Farnesol[SO]	47.76	1694	1695[NIST]	0.08±0	---	---	---
48	Methyl tetradecanoate[Es]	47.88	1698	1699[NIST]	0.07±0	0.01±0	0.03±0	0.035±0
49	alpha-Sinensal[SO]	48.26	1712	1720[PubChem]	0.26±0	---	0.05±0	0.005±0
50	Tetradecanoic acid[Ac]	49.38	1757	1757[NIST]	2.83±0.01	2.65±0.07	4.03±0.13	4.29±0.2
51	Nootkatone[SO]	49.42	1758	1763[NIST]	0.23±0	---	---	---
52	2-Ethylhexyl salicylate[Es]	49.80	1773	1769[NIST]	0.05±0.01	---	0.04±0	0.01±0
53	Methyl n-pentadecanoate[Es]	50.48	1801	1803[PubChem]	0.07±0.02	---	---	---

54	2-Hydroxycyclopentadecanone[C]	51.19	1831	1839,1[NIST]	0.7±0	0.09±0	0.43±0.02	0.09±0
55	Pentadecanoic acid[Ac]	51.59	1848	1851[Pherobase]	1.66±0.06	0.18±0.01	0.74±0.01	0.19±0.03
56	Methyl palmitoleate[Es]	52.27	1877	1881[PubChem]	0.11±0.05	0.13±0	0.23±0.01	0.24±0.01
57	Farnesyl acetone[SO]	52.44	1884	1892[PubChem]	0.2±0	0.03±0	0.04±0	0.06±0.02
58	Methyl palmitate[Es]	52.91	1905	1905[NIST]	3.29±0.03	0.16±0	0.48±0	0.39±0.01
59	Palmitic acid[Ac]	54.92	1997	1997[NIST]	40.71±0.11	54.81±2.64	56.36±0.26	54.79±1.12
60	cis,cis-Linoleic acid[Ac]	55.66	2034	2078[Pherobase]	0.17±0.09	0.03±0	---	---
61	cis-10-Heptadecenoic acid[Ac]	55.72	2037	2073,2[NIST]	0.62±0.02	0.04±0.02	0.29±0.13	0.05±0.01
62	cis-Oleic Acid[Ac]	55.86	2044	2090[NIST]	0.27±0	0.04±0.02	---	---
63	Heptadecanoic acid[Ac]	56.13	2057	2059[PubChem]	0.59±0.03	0.11±0.01	0.10±0.02	0.20±0
64	Methyl linoleate[Es]	56.30	2065	2067[Pherobase]	4.12±0.04	0.03±0.01	0.38±0.02	0.09±0
65	Methyl linolenate[Es]	56.46	2074	2073[PubChem]	1.41±0	0.2±0.02	0.51±0	0.27±0.01
66	Methyl elaidate[Es]	56.54	2078	2075[PubChem]	1.27±0.05	0.21±0	0.04±0	0.03±0
67	Methyl 11-octadecenoate[Es]	56.63	2082	2089[PubChem]	0.41±0.04	---	0.18±0	0.29±0.08
68	alpha-Linolenic acid[Ac]	57.88	2147	2143[PubChem]	27.01±0.13	33.91±1.26	25.9±1.06	29.68±0.14
69	cis-Vaccenic acid[Ac]	58.00	2152	2161,8[PubChem]	1.06±0.03	6.46±1.27	3.25±0.2	5.69±0.08
70	Stearic acid[Ac]	58.17	2161	2161[NIST]	0.23± 0.24	0.09±0.03	0.04±0.01	0.20±0
71	Tributyl acetylcitrate[DAc]	59.20	2216	2224[NIST]	---	0.06± 0	0.09±0	0.01±0
	Total content (%)				**98.69±0.04**	**99.40±0**	**97.09±0.1**	**98.37±0.99**

RT – retention time in minutes; IKc – Kovats indices calculated using a mixture of C_9-C_{26} alkanes; IKl – Kovats indices obtained from the literature; Es – ester; A – alcohol; Ac - carboxylic acid; Al – aldehyde; CA – aromatic compound; H – hydrocarbon; M – monoterpene; MO – oxygenated monoterpene; S – sesquiterpene; SO – oxygenated sesquiterpene; C – ketone; DAc – carboxylic acid derivative. Compounds were identified based on mass spectrometry data and by comparing IKc to IKl; references for IKl: NIST, Pherobase, ChemSpider and PubChem. Quantification was performed using the area normalization technique.

4. Conclusion

When a packaged food product is consumed or thrown away, the packaging is usually discarded, leading to severe environmental damage. Thus, packaging material types and the way we deal with waste must be transformed urgently. This, in turn, has led to growing interest in the development of green-based packaging. This pioneering work shows the viability of using OPF as a mechanical reinforcement and as a bioactive additive in films based on persimmon puree. The films showed high solubility and permeability to water vapor, likely due to the hydrophilic nature of the polymer matrix and glycerol addition. It is important to note that even with the direct addition of flour in the filmogenic solution, the films were homogeneous, demonstrating an adequate dispersion of this residue in the persimmon puree. The incorporation of OPF contributed to the increase in the content of total phenolics and the antioxidant capacity of the persimmon films. In addition, OPF was responsible for most of the identified volatile compounds (97.18%), including those with important aroma and bioactivity properties verified in the films. This means that even when considering the orange peel flour preparation and that of the films by casting at temperatures of at least 60°C, many interesting compounds and their properties were retained. These results suggest that persimmon films incorporated with OPF have the potential to develop active primary food packaging with improved antioxidant and aromatic properties. In addition to bioactivity, considering the other mechanical and barrier characteristics evaluated, some specific uses proposed for these films are as herb and tea sachets for infusion, edible coating for vegan cheeses, and as straws or other components for alcoholic drinks and other cold fruity beverages in order to accentuate the sensory and functional characteristics of these products. Studies like these are aligned with circular economy principles in terms of preventing post-harvest fruit loss and converting organic wastes into new materials to create products with high added value in the context of biorefineries, maximizing the use of resources, and providing positive impacts on the environment.

CRedit AUTHORSHIP CONTRIBUTION STATEMENT

Andreza Salles Barone: Investigation, Writing – original draft, Validation. **Julia Rabelo Vaz Matheus:** Investigation, Writing – original draft. **Mônica Regina da Costa Marques:** Resources, Formal analysis. **Ana Maria Furtado de Souza:** Resources, Formal analysis. **Willian Hermogenes Ferreira:** Resources, Formal analysis. **Ricardo Felipe Alves Moreira:** Investigation, Writing – original draft, Validation, Formal analysis. **Ana Elizabeth Cavalcante Fai:** Investigation, Writing – original draft, Validation, Resources, Funding acquisition, Supervision.

CONFLICT OF INTEREST

The authors declare no conflict of interest.

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CONCLUSÃO GERAL

Se por um lado a pandemia de COVID-19 contribuiu para o aumento do uso e descarte de plásticos convencionais, por outro, alertou para novas oportunidades para a área de embalagens de alimentos. O uso de resíduos agroindustriais para a produção de bioplásticos pode ajudar a reduzir a poluição branca, contribuir para o desenvolvimento de produtos de alto valor agregado e gerar renda adicional. Além disso, o remodelamento do sistema econômico existente aponta para a ascensão da economia circular e por sistemas de embalagens mais circulares, meta essa urgente no contexto do "novo normal" pós COVID-19.

Os resíduos gerados majoritariamente das perdas pós-colheita e durante o processamento industrial de alimentos se mostram como materiais alternativos com grande potencial a ser explorado para o desenvolvimento de bioplásticos e como veículo de entrega de compostos bioativos. Neste estudo, o caqui se mostrou uma matéria prima promissora para a obtenção de filmes ativos antioxidantes e aromáticos com propriedades mecânicas e de barreira satisfatórias para confecção de uma embalagem primária e flexível de alimentos. Além disso, a adição de farinha de casca de laranja contribuiu para melhora na resistência mecânica dos filmes, enquanto o glicerol, como aditivo plastificante, teve influência positiva sobre a elongação na ruptura. A caracterização dos filmes desenvolvidos possibilitou o vislumbre de alguns atributos interessantes desses materiais como componentes de embalagens, destacando-se: flexibilidade, solubilidade, cor vívida e atrativa que reflete a coloração da polpa do caqui maduro, potencial antioxidante e rica composição em compostos voláteis. Sugere-se, assim, sua aplicação como sachês de ervas para influsão e chá onde a embalagem poderia se dissolver junto ao conteúdo e proporcionar o aumento de teor de antioxidantes, além de incrementar as propriedades aromáticas dessas bebidas. Outra proposta inovadora de aplicação seria como embalagem primária em queijos veganos ou em barras de chocolate, de forma a incrementar o conteúdo de bioativos e conferir compostos de sabor/aroma frutados a esses produtos.

Em suma, os filmes desenvolvidos têm potencial como embalagem ativa de alimentos e como materiais biodegradáveis e compostáveis, estudos esses que serão realizados futuramente para comprovação dessas propriedades em adição aos testes de aplicação dos mesmos.