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**FILMES À BASE DE BLENDA AMIDO-PECTINA COM DIFERENTES RESÍDUOS  
AGROINDUSTRIAIS: POTENCIAIS EMBALAGENS INTELIGENTES E ATIVAS  
PARA ALIMENTOS**

Rio de Janeiro

2024



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Tese de Doutorado apresentada ao Programa de Pós-Graduação em Alimentos e Nutrição da Universidade Federal do Estado do Rio de Janeiro, como parte dos requisitos necessários à obtenção do título de Doutora em Alimentos e Nutrição.

Orientadora:

Profa. Dra. Ana Elizabeth Cavalcante Fai

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*Dedico esta Tese de Doutorado ao meu Vô Rabelo,  
minha inspiração de força, persistência e  
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## RESUMO

O objetivo dessa tese foi desenvolver e caracterizar filmes biodegradáveis inteligentes-ativos à base de amido-pectina adicionados de resíduos agroindustriais, utilizando diferentes estratégias de incorporação, e avaliar potencial aplicação em sistemas de embalagens de alimentos. Os resíduos foram coletados e tratados, obtendo-se precipitado liofilizado da manipueira, pó de casca de cenoura roxa, batata doce roxa, beterraba e folhas de oliveira desidratadas em estufa. Os resíduos foram caracterizados e incorporados nas suspensões filmogênicas na forma de pó, exceto para os resíduos de cenoura roxa, também incorporados como extrato. Os filmes foram desenvolvidos por *casting* e caracterizados quanto às propriedades mecânicas, físicas, físico-químicas, colorimétricas, morfológicas, térmicas, de barreira ao vapor de água e radiação UV, atividade antioxidante e propriedades de biodegradação e fitotoxicidade. O potencial de aplicação dos filmes selecionados foi avaliado como selos inteligentes, tampas biodegradáveis, revestimentos e sachês hidrossolúveis comestíveis. Todos os filmes desenvolvidos foram facilmente destacados das placas e apresentaram maleabilidade, ausência de rachaduras ou poros macroscópicos. No entanto, suas propriedades foram influenciadas pela forma de incorporação e tipo de resíduo, além dos diferentes teores de amido e/ou pectina utilizados. O filme com manipueira se apresentou menos uniforme, mais áspero e menos transparente à medida que a concentração do resíduo aumentou na formulação, provavelmente devido à formação de agregados na matriz. Sua adição diminuiu a resistência mecânica e aumentou a flexibilidade e a hidrofiliabilidade sem impacto na solubilidade em água dos filmes. Destaca-se que foram biodegradáveis e não apresentaram componentes tóxicos para a germinação de sementes de feijão. A incorporação do extrato aquoso de pó de casca de cenoura roxa resultou em filmes com maior resistência mecânica, enquanto a adição direta do pó e de seu extrato aquoso com solvente eutético natural profundo (NADES) resultou em filmes com maior sensibilidade colorimétrica à variação de pH e exposição ao vapor de amônia. Os filmes foram eficazes como selos inteligentes na avaliação do frescor de um produto *plant-based* proteico. A incorporação do pó de casca de batata doce roxa e de beterraba resultou em filmes biodegradáveis, não fitotóxicos, com relevante propriedade halocrômica e elevada capacidade de proteção UV. Os filmes adicionados de batata doce roxa apresentaram maior resistência mecânica e sensibilidade colorimétrica à variação de pH e exposição ao vapor de amônia, enquanto aqueles adicionados de beterraba apresentaram maior barreira ao vapor de água, capacidade antioxidante, velocidade de biodegradação e de germinação de sementes de feijão. Ambos os filmes foram promissores como selo inteligente para monitorar o frescor de um alimento *plant-based* e como tampas biodegradáveis, substituindo o PVC, em embalagens de morangos refrigerados. A otimização da formulação dos filmes adicionados de pó de folha de oliveira resultou em um filme com propriedades mecânicas e de opacidade satisfatórias, elevada proteção UV e solubilidade em água, ideal para a produção de sachê hidrossolúvel. A aplicação do filme otimizado como sachê comestível hidrossolúvel foi satisfatória, apresentando rápida solubilização aquosa durante cocção de arroz e preparo de infusão. O resultado da análise sensorial foi promissor, com elevada aceitação global e ótima intenção de compra. Os filmes resultantes demonstraram potencial para serem utilizados como diversas embalagens ativas e inteligentes de alimentos.

**Palavra-chave:** bioplásticos; embalagem primária; casting; subprodutos; bioeconomia circular.

## ABSTRACT

The objective of this thesis was to develop and characterize intelligent-active biodegradable films based on starch-pectin, to which agroindustrial residues were added using different incorporation strategies and to evaluate their potential application in food packaging systems. The residues were collected and treated to produce a freeze-dried cassava precipitate, powdered purple carrot peel, purple sweet potato, beet, and olive leaves. The residues were characterized and incorporated into the film-forming suspensions in powder form, with the exception of the purple carrot residue, which were also incorporated as an extract. The films were developed by casting and characterized for their mechanical, physical, physicochemical, colorimetric, morphological, thermal and barrier properties against water vapor and UV radiation, antioxidant activity, biodegradability and phytotoxicity. The potential application of the selected films was evaluated as intelligent tags, biodegradable lids, coatings and edible water-soluble sachets. All developed films were easily removed from the plates and showed malleability, no cracks or macroscopic pores. However, their properties were influenced by the method of incorporation and the type of residue, as well as by the different starch and/or pectin content. The film with cassava wastewater was less uniform, rougher and less transparent with increasing residue concentration in the formulation, probably due to the formation of aggregates in the matrix. The addition of cassava wastewater reduced the mechanical strength and increased the flexibility and hydrophilicity without affecting the water solubility of the films. It is noteworthy that they were biodegradable and contained no toxic components for the germination of bean seeds. Incorporation of the aqueous extract of the purple carrot peel powder resulted in films with greater mechanical resistance, while direct addition of the powder and its aqueous extract with natural deep eutectic solvent (NADES) resulted in films with greater colorimetric sensitivity to pH variation and exposure to ammonia vapor. The films were effective as an intelligent tag in evaluating the freshness of a plant-based protein product. The incorporation of purple sweet potato and beet peel powder resulted in biodegradable, non-phytotoxic films with relevant halochromic properties and high UV-protective capacity. The films containing purple sweet potato showed higher mechanical strength and colorimetric sensitivity to pH variation and ammonia vapor, while the films containing beet showed higher barrier to water vapor, higher antioxidant capacity, biodegradation speed, and higher bean seed germination capacity. Both films showed promise as intelligent tags for monitoring the freshness of plant-based foods and as biodegradable lids replacing PVC in refrigerated strawberry packaging. The optimization of the film formulation with the addition of olive leaf powder resulted in a film with satisfactory mechanical and opacity properties, high UV protection and water solubility, ideal for the production of water-soluble sachets. The application of the optimized film as a water-soluble edible sachet was satisfactory, showing rapid aqueous dissolution during rice cooking and infusion preparation. The result of the sensory analysis was promising, with high global acceptance and good purchase intent. The resulting films have the potential to be used as various active and intelligent food packaging.

**Keyword:** bioplastics; primary packaging; casting; by-products; circular bioeconomy.

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## 1. INTRODUÇÃO

O consumo excessivo de recursos naturais e a produção material elevada, em especial de plásticos, são alguns dos maiores desafios contemporâneos. As atividades humanas estão provocando mudanças atmosféricas, geológicas, hidrológicas e biosféricas. Essa influência humana no planeta é o que caracteriza o que muitos pesquisadores consideram como a atual era geológica denominada de Antropoceno (Velenturf et al., 2021; Willett et al., 2019). Nesse sentido, algumas das atividades que apresentam elevado impacto ambiental negativo são a produção, perda e desperdício de alimentos (Fanzo et al., 2020; Willett et al., 2019) e o consumo em larga escala de plásticos de origem fóssil (Acquavia et al., 2021).

A indústria de alimentos tem sido fundamental para viabilizar a alimentação e nutrição de uma população mundial crescente, envolvendo desde agricultura, processamento de alimentos e embalagem até transporte, vendas e atendimento ao consumidor (Şimşek et al., 2024). É imprescindível um sistema de produção de alimentos robusto capaz de suprir a demanda alimentar mundial crescente, e de contribuir para a diversificação, preservação e distribuição de alimentos com o apoio da ciência e tecnologia de alimentos (Buckle, 2015). As projeções apontam 10 bilhões de pessoas em 2050 (Willett, et al., 2019; Jurgilevich et al., 2016), tornando cada vez mais urgente a implementação de soluções que foquem em um sistema alimentar com uso de recursos com sustentabilidade ambiental, social e econômica, com garantia da segurança alimentar para todos (Şimşek et al., 2024). Atualmente, a cadeia de alimentos é responsável por cerca de 30% das emissões globais de gases de efeito estufa e 70% do consumo de água doce. Além disso, agricultura e pastagem ocupam cerca de 40% da superfície terrestre livre de gelo (Fanzo et al., 2020; Willett et al., 2019). A expressiva perda global de alimentos e geração de resíduos agroindustriais ao longo da cadeia de abastecimento de alimentos também contribuem para impactos negativos no ambiente. Estima-se que o montante de alimentos perdidos represente cerca de 8% das emissões globais de gases de efeito estufa (FAO, 2015), além de ocupar 0,9 milhão de hectares de solo e consumir 306 km<sup>3</sup> de água (FAO, 2014).

Somado a esse cenário, outro contribuinte aos impactos ambientais negativos é o crescente consumo e descarte de plásticos de origem petroquímica (Acquavia et al., 2021). Em 2019, cerca de 370 milhões de toneladas de plásticos foram produzidas no mundo (Plasticseurope, 2020). Dentre todos os plásticos produzidos, cerca de 36% são destinados à

elaboração de embalagens plásticas (UNEP, 2018). Cerca de 50% dos resíduos plásticos descartados são constituídos por produtos plásticos de utilização única, que são rapidamente descartados (Shashoua et al., 2024). Considerando a tendência de produção de plástico ao longo dos anos, projeta-se que, em 2050, serão mais de 25 bilhões de toneladas de plásticos produzidos, cerca de 12 bilhões de toneladas de resíduos plásticos acumuladas em aterros ou ambientes naturais (Geyer et al., 2017), além de um aumento das emissões para 6,5 Gt CO<sub>2</sub>eq (Zheng, Suh, 2019). Menos de 10% dos resíduos plásticos são reciclados expondo-os ao ambiente natural e sujeitando-os à fragmentação até formação de micro e nanoplásticos persistentes no ambiente. Em outras palavras, os plásticos não desaparecem, apenas diminuem de tamanho. A presença desses microplásticos no ar, na água, nos alimentos e em diferentes órgãos do corpo vem se tornando uma preocupação emergente de saúde pública (Shashoua et al., 2024).

Considerando esses aspectos, aumenta a preocupação direcionada em promover impactos positivos que colaborem para reconstituir o meio ambiente e melhorar a qualidade de vida de forma geral, incentivando os diferentes atores sociais a repensar o modelo tradicional de “descarte” associados à economia linear (Shogren et al., 2019). Em contraponto, a economia circular como um novo modelo econômico e de desenvolvimento sustentável ganha visibilidade. A economia circular, inspirada nos conceitos cíclicos inerentes a natureza, propõe um sistema de ciclos de reaproveitamento a fim de esgotar a circularidade dos produtos, componentes e/ou materiais, extraíndo todo seu valor e otimizando sua utilização entre ciclos técnicos e biológicos, colaborando para um consumo mais sustentável, com menor emissão de gases do efeito estufa e geração de resíduos (Murray et al., 2017; Jurgilevich et al., 2016). Assim, a indústria de embalagens desempenha um papel crucial nessa mudança de paradigma uma vez que continua a depender majoritariamente de materiais não sustentáveis, como plásticos de origem fóssil de uso único e embalagens multicamadas, que representam cerca de 15-20% de resíduos sólidos em diferentes países (Tako et al., 2021; Hall, 2017; Tencati et al., 2016).

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 a partir de excedentes vegetais e resíduos agroindustriais, principalmente para o segmento de embalagens de alimentos (Matheus et al., 2021; Meys et al., 2020; Luttenberger, 2019). De modo geral, diversos consumidores estão dispostos a escolher

embalagens sustentáveis, mesmo apresentando maior preço de mercado que os plásticos convencionais (Shen et al., 2020). Nessa circunstância, a crescente busca por polímeros ambientalmente amigáveis aliada à necessidade de aproveitar melhor os resíduos agroindustriais contribuem para o interesse no desenvolvimento de filmes biodegradáveis (Acquavia et al., 2021). A formação de filmes a partir de resíduos alimentares vegetais ocorre devido à presença de polímeros naturais nessas matrizes, tais como amido, gelatina/proteína e compostos lignocelulósicos (Acquavia et al., 2021; Brito, Ferreira & Fai, 2020), sendo o amido um dos biopolímeros mais representativos para o mercado de bioplásticos (Shen et al., 2020).

O amido é considerado um dos biopolímeros mais adequados para a produção de filmes biodegradáveis uma vez que são naturalmente abundantes em diferentes espécies vegetais como tubérculos, leguminosas, cereais e resíduos de plantas agroindustriais, além de serem renováveis, biodegradáveis, de baixo custo e geralmente reconhecidos como seguro para embalar produtos alimentícios. A viabilidade do uso do amido para o desenvolvimento de material termoplástico com propriedades adequadas para a produção de filmes ocorre devido às suas características estruturais e de cristalinidade. Além disso, tanto o amido isolado ou em misturas biopoliméricas constituem-se como um material emergente para embalagens de alimentos com uma produção simples, propriedades funcionais desejáveis e alta taxa de degradabilidade (Su et al., 2022; Bayram et al., 2021). Além do amido, a pectina é polissacarídeo complexo que vem sendo uma opção de matéria-prima para desenvolvimento de filmes biopoliméricos, dado sua abundância, biodegradabilidade, comestibilidade e propriedades físico-químicas que favorecem a formação de filmes com alta flexibilidade, solubilidade em água e propriedade de barreira de umidade (Roy et al., 2023; Bayram et al., 2021). A pectina pode ser extraída a partir de resíduos de biomassa, sendo as cascas de frutas cítricas a principal fonte de obtenção de pectina, especialmente em escala industrial (Mellinas et al., 2020).

Ademais, o uso desses resíduos ricos em componentes com atividade antioxidante, na formulação de filmes biodegradáveis, pode expandir sua aplicabilidade para a produção de embalagens ativas de alimentos. Estas podem atuar como veículo para carrear compostos bioativos que possivelmente irão interagir com o alimento embalado, incrementando sua qualidade nutricional, microbiológica ou físico-química e, assim, prolongando a sua vida útil (Soofi et al., 2021; Bhardwaj, Alam & Talwar, 2019). Assim, o desenvolvimento de embalagens ativas que interagem com os alimentos trazem benefícios adicionais à proteção oferecida pelas

embalagens tradicionais feitas de materiais mais inertes (Bayram et al., 2021; Bhargava et al., 2020). Além das embalagens ativas, existem as embalagens inteligentes que podem fornecer aos consumidores informações sobre as condições ambientais dos alimentos, como temperatura, atmosfera da embalagem, processos de amadurecimento ou deterioração, informando-os sobre o real estado de comestibilidade e validade (Bayram et al., 2021; Bhargava et al., 2020). Os indicadores colorimétricos são comumente pesquisados em sistemas de embalagem inteligente, pois fornecem informações qualitativas ou semiquantitativas a partir de uma interpretação direta a olho nu (Saliu et al., 2018). Diversos pigmentos naturalmente presentes em resíduos agroindustriais vêm sendo pesquisados como indicadores colorimétricos em filmes inteligentes devido às suas propriedades halocrômicas, destacando-se as antocianinas e betalaínas como principais pigmentos incorporados em filmes inteligentes (Bayram et al., 2021; Bhargava et al., 2020).

Assim, os resíduos agroindustriais vegetais, como cascas, sementes, talos e bagaços, constituem-se como fontes de compostos bioativos que podem ser incorporados na matriz polimérica de filmes para atuarem como agentes antimicrobianos, antioxidantes, aromáticos e/ou indicativos (de segurança e qualidade) (Bhargava et al., 2020). Estudos avaliaram o efeito da incorporação de resíduos agroindustriais em filmes à base de amido, tais como farinha de casca de feijoa (Sganzerla et al., 2020), extrato de semente de jaca, casca de uvas negras sem sementes e pó de folhas de moringa (Costa et al., 2021), resíduos de mamão (cascas, sementes e caule foliar) e resíduos secos de chá verde (Sethulakshmi & Saravanakumar, 2024). Além disso, diferentes pesquisas contendo resíduos foram usados para o desenvolvimento de filmes inteligentes indicadores colorimétricos sensíveis ao pH, tais como filmes à base de pectina de casca de melancia usando extrato de beterraba (Guo et al., 2021), filmes à base de amido/quitosana incorporado de extrato de repolho roxo (Rai et al., 2023) e filmes à base de amido incorporado de pó de blueberry (Luchese et al., 2017). Esses estudos reforçam a viabilidade de aproveitar os resíduos agroindustriais vegetais que seriam descartados para a produção de filmes com potencial característica de biodegradabilidade.

A água residuária da mandioca (manipueira) é outro exemplo de resíduo amplamente gerado durante o processamento da mandioca, principalmente para a obtenção da farinha (Oghenejoboh et al., 2021; Cruz et al., 2021), variando entre 300 a 600 L de água residuária por tonelada de raiz processada (Costa et al., 2022; Oghenejoboh et al., 2021). O aproveitamento da manipueira pode contribuir para o melhor manejo desse resíduo, uma vez que reduz os riscos

ambientais do seu descarte inadequado (Costa et al., 2022; Sánchez et al., 2017; Schmidt et al., 2023), além de transformar um subproduto que seria descartado em um produto de alto valor agregado com importância econômica e social (Santos et al., 2022). Considerando a presença de biopolímeros na manipueira, como carboidratos e proteínas (Costa et al., 2022; de Souza, 2021; Schmidt et al., 2023), esses resíduos apresentam uma oportunidade para o desenvolvimento de filmes biocompósitos, fortalecendo uma relevante cadeia produtiva nacional, uma vez que a mandioca é uma importante cultura básica brasileira, produzida principalmente pela agricultura familiar (Cruz et al., 2021).

O cultivo de oliveiras nas últimas duas décadas é crescente tanto no mundo, quanto no Brasil (FAOSTAT, 2024), porém a geração de resíduos é elevada, especialmente folhas, podendo resultar em mais de 18 milhões de toneladas (Martínez-Navarro et al., 2022, 2023). Esse é um resíduo rico em fibras (Fodil et al., 2024) e compostos bioativos (El Adnany et al., 2024), com potencial aproveitamento para o desenvolvimento de embalagem de alimentos (Selim et al., 2022). Algumas pesquisas avaliaram a incorporação deste resíduo em suspensões filmogênicas, majoritariamente sob a forma de extrato (da Rosa et al., 2020; Martiny, Pacheco, et al., 2020; Musella et al., 2021). Assim, a incorporação direta do pó na suspensão filmogênica apresenta um caráter inovador e pode contribuir para um aproveitamento de maiores quantidades de resíduos, além de transportar compostos bioativos e macromoléculas, melhorando as propriedades dos filmes.

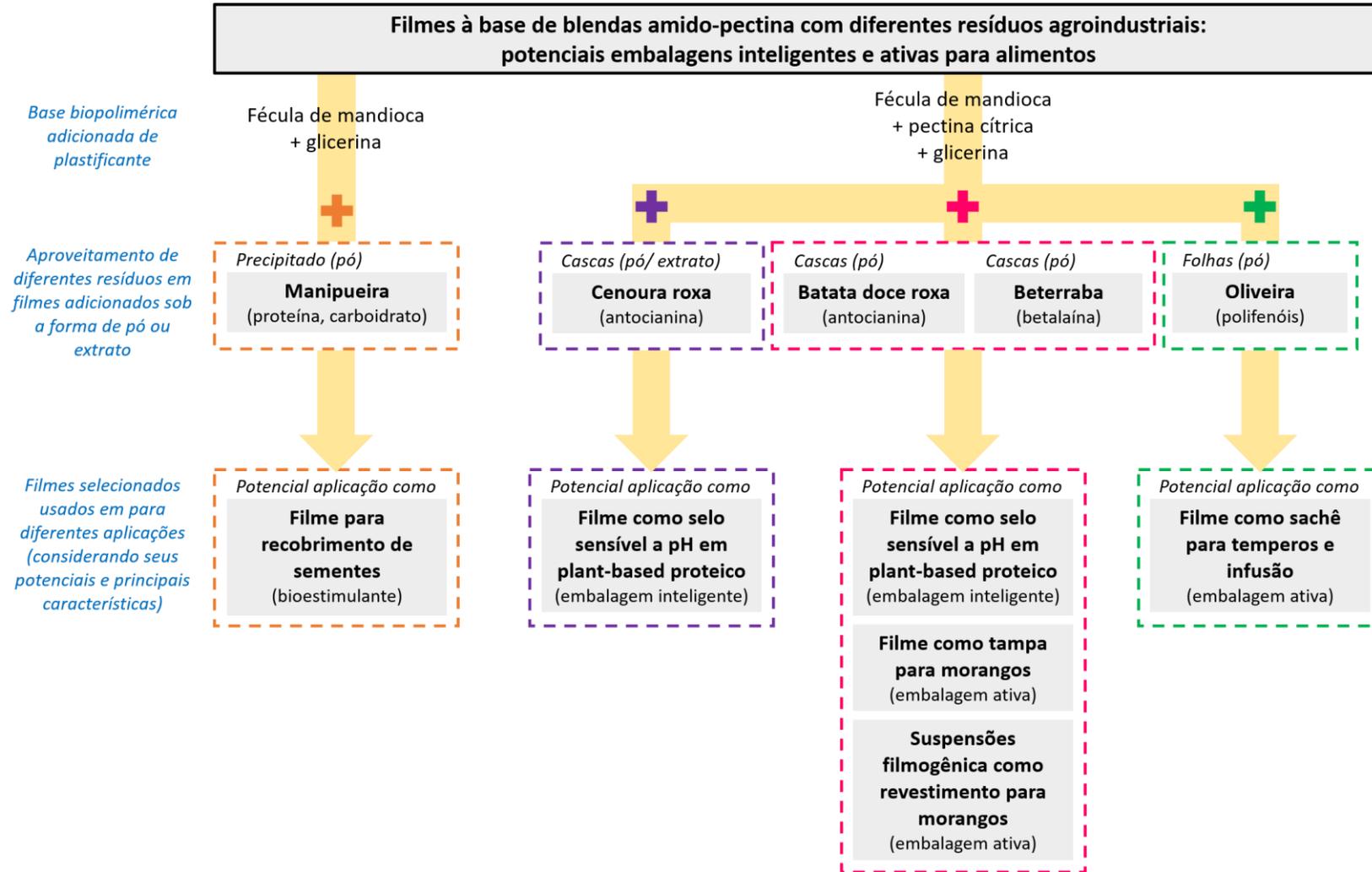
Os resíduos de vegetais roxos, devido à presença de alguns pigmentos bioativos como antocianina (presente na cenoura roxa e batata doce roxa) e betalaína (presente na beterraba), podem não só conferir bioatividade aos filmes, mas também viabilizar tecnologicamente a propriedade de sensibilidade colorimétrica frente à variação das condições ambientais da embalagem decorrentes de processos de deterioração dos alimentos (Bayram et al., 2021; Bhargava et al., 2020).

Considerando o exposto, evidencia-se a importância da valorização e uso estratégico de resíduos agroindustriais, tais como manipueira, cascas dos vegetais roxos (cenoura roxa, batata doce roxa e beterraba) e folhas de oliveira, para o desenvolvimento de filmes biodegradáveis para aplicação como embalagens de alimentos (Figura 1).

Esta tese é apresentada em formato de artigos e trabalhos científicos divididos em oito capítulos. Os quatro primeiros capítulos são compostos por trabalhos de revisão acerca do tema da tese e os quatro últimos capítulos são artigos de pesquisa sobre desenvolvimento de filmes

incorporados de resíduos agroindustriais. São eles: (I) “*A holistic approach to sustainable food waste management and residue utilization*” (publicado no livro *Engineering Aspects of Food Quality and Safety*, Food Engineering Series, Springer em 22 de agosto de 2023); (II) “*Biopolymers as green-based food packaging materials: a focus on modified and unmodified starch-based films*” (publicado na revista *Comprehensive Reviews in Food Science and Food Safety* em 29 de janeiro de 2023); (III) “*Cassava starch films for food packaging: trends over the last decade and future research*” (publicado na revista *International Journal of Biological Macromolecules* em 15 de janeiro de 2023); (IV) “*Starch-based films containing purple halochromic pigments: smart food packaging*” (submetido para a revista *International Journal of Biological Macromolecules*); (V) “*Closing of cycle: cassava starch and wastewater films as biostimulant for bean seed and plant growth enhancement*” (submetido para a revista *Biocatalysis and Agricultural Biotechnology*); (VI) “*Valorization of purple carrot peel as pH-indicating smart tag for plant-based food freshness*” (submetido para a revista *Food Hydrocolloids*); (VII) “*Starch/pectin materials with beetroot or purple sweet potato peels: from chemical properties to application as smart food packaging*” (a ser submetido para a revista *Journal of Agricultural and Food Chemistry*); e (VIII) “*Olive leaves addition on starch-pectin films: optimization, characterization, and evaluation as edible hydrosoluble sachets*” (a ser submetido para a revista *Food Hydrocolloids*).

**Figura 1.** Esquema geral do aproveitamento de diferentes resíduos para o desenvolvimento de filmes biodegradáveis.



## 2. OBJETIVOS

### 2.1. Objetivo geral

Desenvolver e caracterizar filmes biodegradáveis inteligentes-ativos à base de amido-pectina adicionados de resíduos agroindustriais (manipueira, cascas de vegetais roxos e folhas de oliveira), utilizando diferentes estratégias de incorporação, e avaliar o potencial de aplicação em sistemas de embalagens de alimentos.

### 2.2. Objetivos específicos

- (a) Caracterizar a manipueira e desenvolver diferentes formulações de filmes à base de amido com incorporação de manipueira e caracterizá-los;
- (b) Caracterizar as cascas de vegetais roxos (cenoura roxa, batata doce roxa e beterraba), desenvolver diferentes formulações de filmes à base de amido-pectina com incorporação de pó ou extratos dos resíduos de vegetais roxos, caracterizar e aplicar como selos inteligentes em *plant-based* análogo ao frango (filmes incorporados de cascas de cenoura roxa, batata doce roxa e beterraba) e como embalagem ativa (filmes como tampas e suspensões filmogênicas revestimentos incorporadas de cascas de batata doce roxa e beterraba) em morangos refrigerados;
- (c) Caracterizar as folhas de oliveira, desenvolver filme à base de amido-pectina com incorporação de folhas de oliveira otimizando formulação com planejamento experimental, caracterizar os filmes, aplicar o filme otimizado como sachê hidrossolúvel comestível para temperos ou para infusão de pó de folhas de oliveira e analisar sensorialmente três preparações: arroz branco coccionado com adição do sachê hidrossolúvel comestível contendo dois tipos de temperos e infusão de pó de folhas de oliveira elaborada a partir do sachê hidrossolúvel comestível.

## **CAPÍTULO I**

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*Publicado*

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**A HOLISTIC APPROACH TO SUSTAINABLE FOOD WASTE MANAGEMENT  
AND RESIDUE UTILIZATION**

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## **ABSTRACT**

Waste generation is inherent to food production. There are several strategies that can be implemented to decrease the amount of waste generated, but good management and intelligent use of such waste are essential. Additionally, surplus vegetable production in relation to consumption/processing also becomes waste at the end of the food chain and thus attention must be given to reducing these losses. The transformation of residues and vegetables - that would otherwise be lost - into new products with maximum added value is aligned with the precepts of the circular economy and the proposals of the UN. Scenarios like the one experienced during the COVID-19 pandemic alert us to the fact that changes towards a more ecologically healthy environment are imperative and that the world can - and should - be cleaner and greener. There are several studies on the use of agro-industrial residues, but it is important to increase the scope of such studies, interrelating many other aspects that have a direct impact on the amount, type, and management of the residues generated. Thus, a holistic approach to sustainable food waste management and residue utilization is proposed and discussed in this chapter.

**Keywords:** Bioeconomy; Economic aspects; Sociocultural aspect; Sustainability; Nutritional impact; Food system.

## 1. INTRODUCTION

Worldwide food waste reaches 1.3 billion tons annually throughout the entire food supply chain from farm to fork. This has not only caused environmental and health impacts but has also led to an economic crisis (Arun et al., 2020; Teigiserova et al., 2019a).

It is important to highlight that in addition to the problems related to food loss, food processing results in the generation of inedible food residue that, despite the efforts being made to mitigate quantities, will persist. The good news: these residues may represent a stable renewable material for the future biobased value chains and products (Teigiserova et al., 2019a).

This is possible due to the chemical composition and bioactive molecules found in food residues. If food residues are used to recover macro and micronutrients, bioactives, biopolymers, among others, it is feasible to reduce the cost of these valuable compounds as well as to reduce waste (Arun et al., 2020). Thus, advanced methodologies to characterize, extract and transform those compounds through technology and biotechnology routes have been explored in the last years. Obtaining new sustainable materials for different industrial purposes has also been extensively investigated (Brito et al., 2021, 2020a, 2020b; Matheus et al., 2021, 2020b; Teigiserova et al., 2019a).

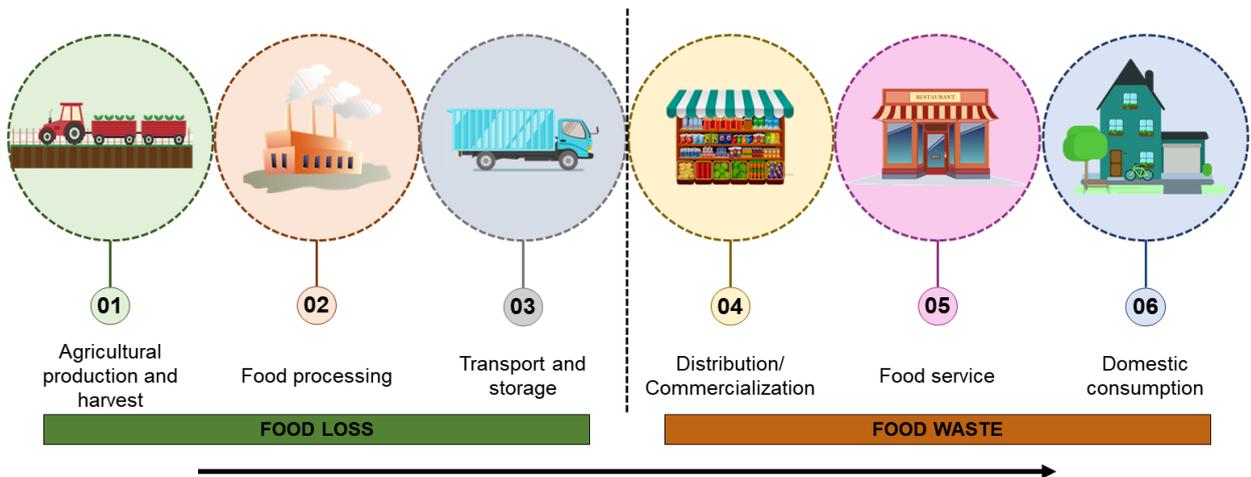
In light of this situation, a holistic approach is urgently desirable to reduce food loss and waste, as well as for the management of food residue inherent in food production. Additionally, technological and biotechnological research is required to maximize the economic incomes, by transforming food residues into valuable bioproducts, with low-cost investments.

The sustainable utilization of food residues is aligned with the precepts established in the circular economy, which adopts the concept of re-using and recycling biowastes, leading interested parties and producers to embrace more environmentally-friendly processes (Ng et al., 2020). The circular bioeconomy adopts the use of renewable biological resources to transform them into high-value products, such as bioactive components, bioplastic, and bioenergy, and to preserve the resources for a longer period of time with the objective of producing zero waste and reducing greenhouse gases (GHG) emissions (Sharma et al., 2021). This model is also aligned with the Sustainable Development Goal (SDG) Target 12.3, which aims to halve global food residues at the retail and consumer levels and reduce food losses, including postharvest losses, along supply chains by 2030, by ensuring sustainable consumption and production patterns (Lipinski, 2020).

Thus, a holistic approach to sustainable food waste management and residue utilization is proposed in this chapter.

## 2. ECONOMIC AND NUTRITIONAL ASPECTS OF FOOD LOSS AND WASTE: AN OLD ISSUE WITH MODERN RELEVANCE

Food supply chains include primary agricultural production, manufacturing, retail, and household consumption. Throughout its life cycle, food may be lost or wasted due to technological, economic and/or social reasons (Otles et al., 2015). Food loss or waste is defined as the masses of food lost or wasted in some part of the food chain between the producer and consumer, from edible products intended for human consumption (FAO, 2011). Food loss is an unintended loss in edible food quantity or quality before consumption during harvest, post-harvest handling, processing, and distribution. Food waste is when safe and nutritious food for human consumption is discarded or not consumed, primarily in the retail and consumption stages (including services) (Figure 1) (de Brito Nogueira et al., 2020; Teigiserova et al., 2019b; FAO, 2011). Table 1 shows several reasons for food loss and waste at the distinct stages of the food supply chain.



**Figure 1.** Food loss and food waste: where each of them is in the stages of the food supply chain.

**Table 1.** Possible causes of food loss and waste throughout the food supply chain.

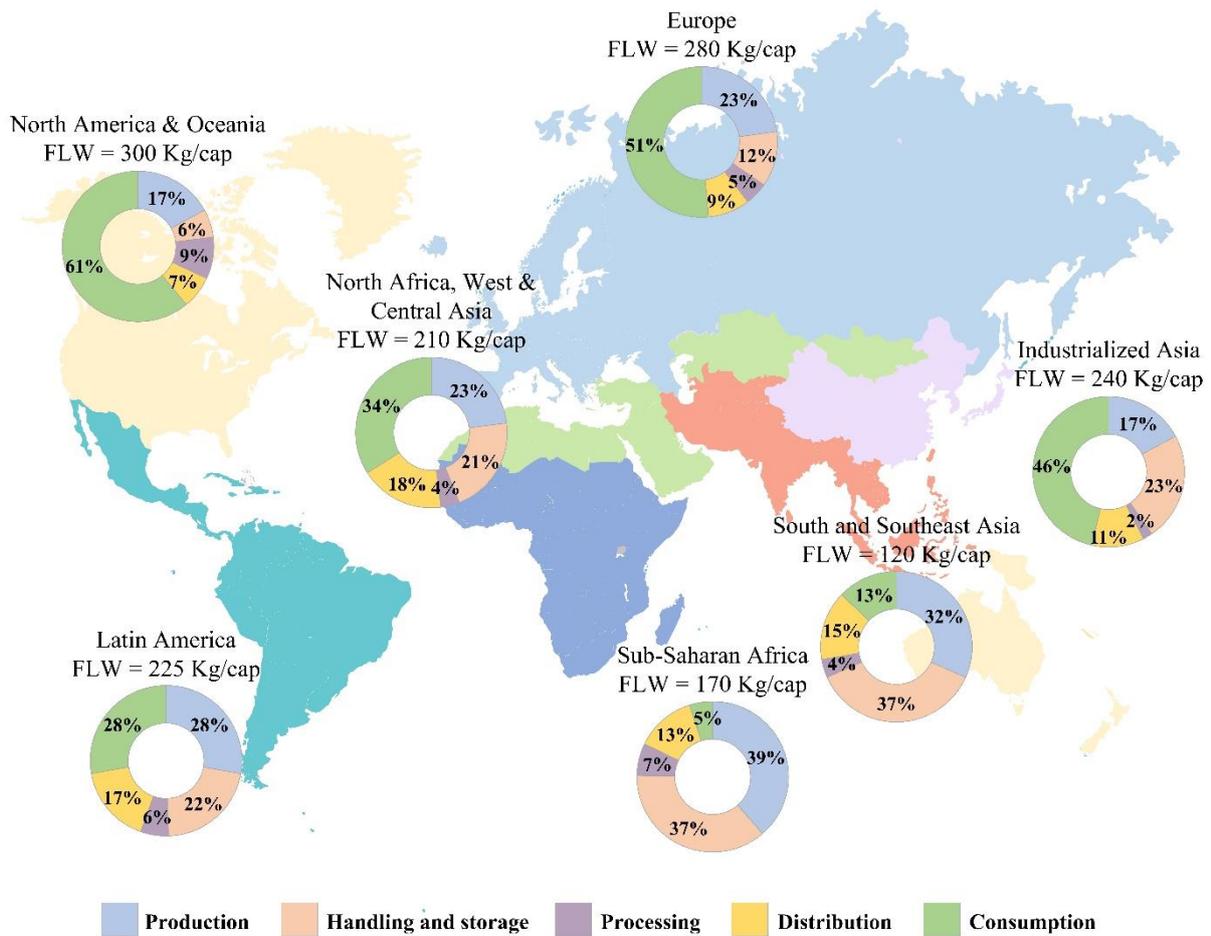
<b>Stage of Food Supply Chain</b>	<b>Causes of food loss and waste</b>
Production	Infrastructural limitation Overproduction Harvesting timing and method (manual/mechanical) Pesticides and fertilizers Economic problems Quality standards and norms
Postharvest handling and storage	Degradation and spoilage of product composition Loss during transportation from farm to distribution Storage infrastructure
Processing and packaging	Unavoidable losses Technical malfunctions Methods and changes in processing lines Contamination in processing lines Legislation restrictions Packaging system Overproduction
Distribution and marketing	Inappropriate transport conditions (temperature-controlled aircrafts and ships) Contamination of transportation Transportation and market facilities Road and distribution vehicles Packaging management Commercial conditions Consumer preference
Consumption	Composition unit and size of household Income group Demographics and culture Individual attitude Cooking practices and methods

Adapted from: Singh, 2020

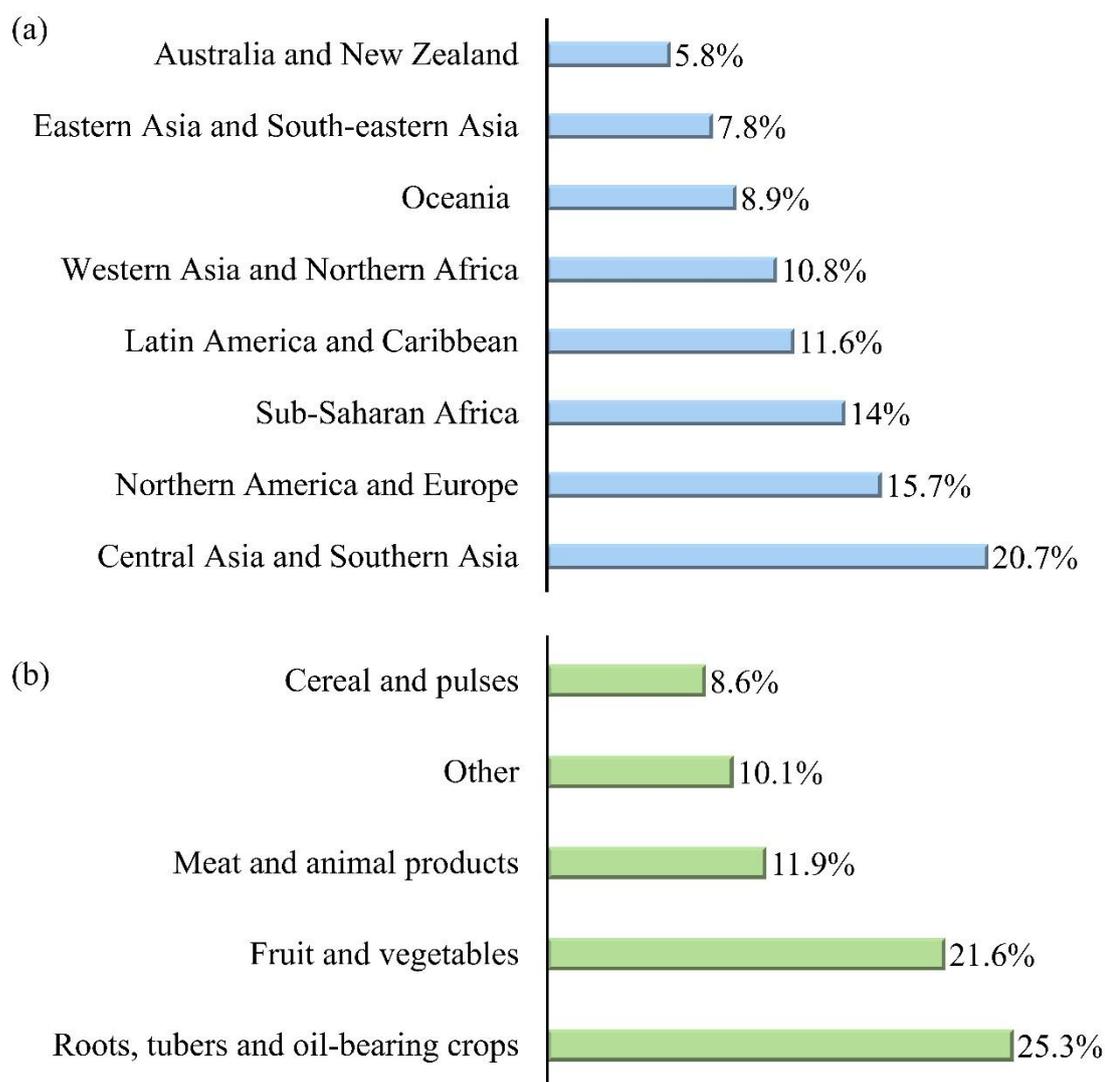
Food waste constitutes a main driver of both economic and environmental degradation, since natural resources are depleted during the production, preparation and disposal stages (Wakefield and Axon, 2020). From an economic point of view, unsustainable food production leads to negative impacts on the supply chain, including higher prices, increased price volatility, and decreased profits and economic value of the food itself (Roodhuyzen et al., 2017). The loss of food represents a loss of significant amounts of money and other resources, such as investments in the supply chain in food production, including fresh water, labor, energy, agricultural chemicals, and other inputs for food production, when the intended purpose of

feeding people is not fulfilled (Buzby et al., 2014). It is estimated that 1.3 billion tons per year of edible food produced for human consumption is lost or wasted. This represents about one-third of the edible food produced during the food supply chain (FAO, 2011). A more recent study estimates that about 14% of food is lost in the pre-retail levels (e.g., agriculture, post-harvest, slaughter, and catch) (FAO, 2019). Regarding the global economy, the value of food lost or wasted annually is estimated at US\$ 1 trillion (FAO, 2015a). The economic cost of waste management comprises the maintenance of landfills, transportation, treatment plant operation and separation and segregation of the waste. The total annual economic, environmental and social cost of food waste for the global economy considering food waste that is not collected separately and disposed of in landfills is US\$ 2.6 trillion (Singh, 2020).

The quantity of food loss and waste is influenced by several drivers, including the modernization of society, increased globalization of trade, urbanization, cultural changes, dietary transitions, and sociodemographic factors (Wakefield and Axon, 2020). Regarding per capita income, developed countries waste six times more food by weight than developing countries (Chen et al., 2020). However, food losses in high-income and low-income nations are estimated to be on the same level. In developed countries, more than 40% of food is lost during the marketing and consumption stages, while in developing countries about 40% is lost during the postharvest and processing stages (FAO, 2011). Figure 2 presents the quantity of worldwide food loss and waste generation and its distribution along the food supply chain, based on data reported by FAO (2011) and by Lipinski et al. (2013). At the regional level, food loss ranges from 5.8% in Australia and New Zealand, to 20.7% in Central and Southern Asia (Figure 3 a) (FAO, 2019). Food commodities, roots, tubers and oil-bearing crops report the highest losses – 25.3%, followed by fruit and vegetables – 21.6%, mostly owing to their highly perishable nature (Figure 3 b) (FAO, 2019).



**Figure 2.** Worldwide food loss and waste and its distribution in the different stages (production; handling and storage; processing; distribution; and consumption) of the food supply chain in 2009. FLW refers to the total food loss and waste in kg per capita/ year.



**Figure 3.** Share of global food loss in 2016 by (a) region and (b) commodity from the farm to, but excluding, retail stage.

It is important to add the nutritional panorama that has been drawn to this economic data. This outlook portrays a global food system that is failing to meet the nutritional needs of the world population, and focuses on health issues related to insufficient micronutrient consumption (Ritchie et al., 2018). Food insecurity, defined as “inefficient access to safe, sufficient and nutritious food to meet a person's dietary needs and food preferences in order to pursue an active and healthy lifestyle” (Tester et al., 2020) is a worldwide public health problem. This concept has been associated with hunger, malnutrition, obesity, and inadequate access to nutritionally satisfying foods (Brown et al., 2019).

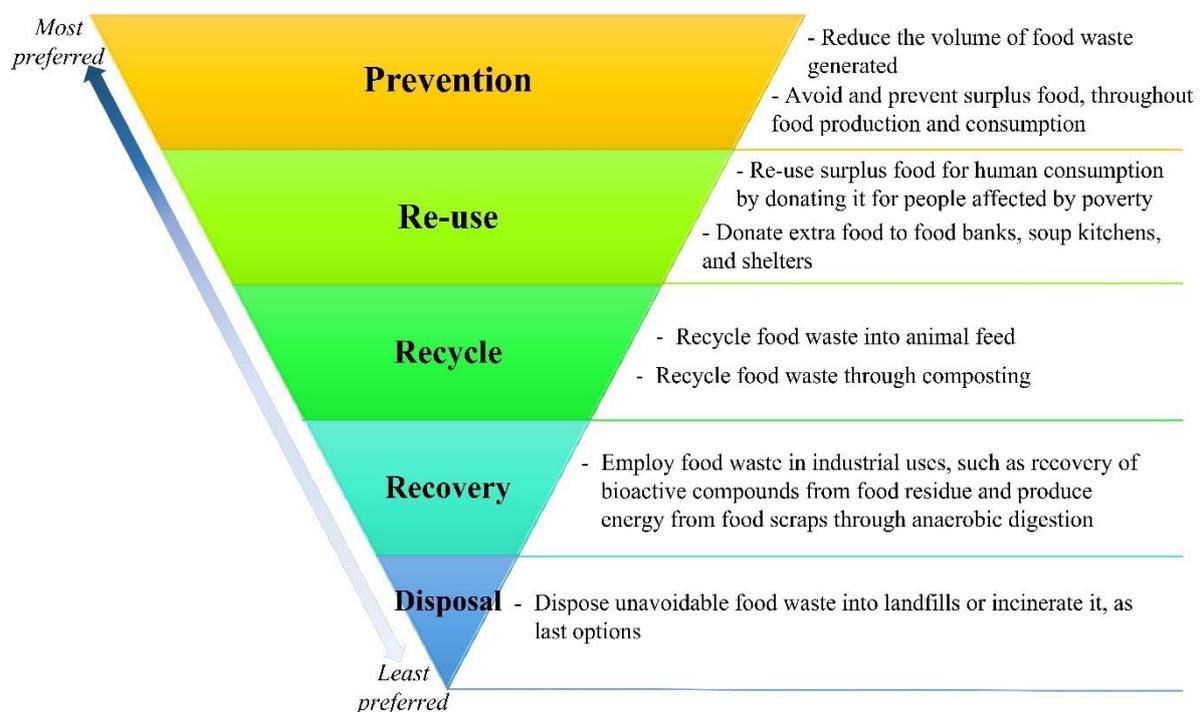
Although food insecurity can be obesogenic, linked to unhealthy eating patterns, it can also lead to weight loss, especially in its most severe form – hunger (Morales and Berkowitz, 2016). It is estimated that in 2020 between 720 and 811 million people suffered from hunger all over the world (FAO et al., 2021), contributing to the failure to achieve the UN's Sustainable Development Goals (SDG), in particular SDG2, which aims to eradicate hunger, achieve food security, improve nutrition and promote sustainable agriculture by 2030 (UN, 2020). This fact is observed and reported mainly in low- and middle-income countries, where the population does not have regular access to safe, nutritious and sufficient food (FAO et al., 2019), accompanied by the disincentive of public and social policies that contribute to addressing the problem (Watson et al., 2020). Food insecurity may also be associated to dietary patterns characterized by a lower consumption of healthy food groups and a poor quality of diet, particularly regarding the intake of fruit and vegetables (Morales and Berkowitz, 2016). In addition, it is known that in low-income neighborhoods, there is a dichotomy between the limited supply of fresh and nutritious food and an increase in fast-food outlets, which are “low cost” per calorie compared to healthier foods (Tester et al., 2020). Understanding these relationships and the social impacts linked to them is particularly useful, requiring a profound overhaul of food systems in order to provide healthy and sustainably produced diets for a constantly growing world population (FAO et al., 2019; Torres-León et al., 2018).

If, on the one hand, there is scarcity and a lack of access to nutritionally healthy food, on the other, we find the problem of waste generation and mismanagement inherent in food production (Lai et al., 2017). It is estimated that about 1.3 billion tons of food intended for human consumption is wasted each year (Laso et al., 2021) and that on a global scale the nutritional cost resulting from these losses would feed approximately 940 million people around the world (Abbade, 2020).

In addition to these drivers, the world is also currently facing the COVID-19 pandemic, with high potential to exacerbate worldwide malnutrition (Kurtz et al., 2021), which has increased the urgency to solve the problem of food loss and waste, as food systems have been trying to respond to sudden changes in demand, scarcity of jobs, and lower available incomes (Lipinski, 2020). The pandemic has led to higher waste generation in the consumer stage mainly owing to the over-buying trend and improper storage of high quantities of foods, as well as a disruption in food supply chains, due to road closures which generated an accumulation of products, resulting in increased levels of food loss and waste (Boyacı-Gündüz et al., 2021).

Aldaco et al. (2020) reported that COVID-19 has had a low impact on the overall food loss and waste generation, however it resulted in a 12% higher generation of food waste at the household level in Spain.

To overcome the surplus generation of edible food and consequently the production of food waste, a food waste hierarchy is proposed to reduce and recover food excess, by separating and managing it. Reducing and recovering surplus food may result in both economic and environmental benefits (EPA, 2012). The food recovery hierarchy establishes a hierarchical order between the different ways to reduce food waste, as follows: first, prevention, by reducing surplus at the source; second, recovery, by reusing for human consumption; and third, recycling, by feeding animals, creating energy or compost (Mourad, 2016). From an economic point of view, preventing surplus food may save money since disposal costs would fall; a decrease in costs related to sewage and electricity treatment; decrease in purchase costs, since only what is needed is purchased; increased tax deductions for food donations to charities; and increased revenue from the sale of compost made of food scraps (EPA, 2012). Figure 4 shows the food waste hierarchy.



**Figure 4.** Food waste hierarchy.

Traditionally, agricultural and food waste management included incineration, landfills, composting and application as animal feed. Currently, most of the food residues end up in landfills, which leads to environmental and health concerns, e.g., emission of GHG, bacterial contaminations, and infectious diseases (Ng et al., 2020). However, to achieve more sustainable processes, and decrease costs while increasing the effectiveness of processes, industries are looking for innovative strategies (Kavitha et al., 2020), such as an approach for biofuel and biofertilizer production, besides energy recovery from food waste using ultra-fast hydrolysis and co-digestion process, making possible zero-food waste disposal (Ma and Liu, 2019).

### **3. ENVIRONMENTAL ASPECTS: GOING BEYOND REDUCING FOOD LOSS AND WASTE, A PERSPECTIVE ON AGROINDUSTRIAL RESIDUE UTILIZATION**

Food production is currently one of the main causes of global environmental change. Degradation occurs due to the high consumption of water, land, and energy, generation of GHG (e.g., carbon dioxide, methane, and nitrous oxide), and a loss of biodiversity (FAO, 2013; Gustavsson et al., 2011; Willett et al., 2019). The impacts of the food production system include the emission of about 30 - 35% of all GHG emitted in the world (Foley et al., 2011). Considering the indirect emissions associated with the change in land cover, agricultural production alone is responsible for approximately 86% of the total emissions from the food system (Vermeulen et al., 2012). Agriculture also consumes about 70% of the planet's freshwater (Foley et al., 2011; Khan and Hanjra, 2009) and about 85% of the world's water consumption (Pfister et al., 2011). Furthermore, cultivation and pasture areas already occupy an average of 40% of the land's surface (Foley et al., 2005). These data reinforce the need to transform the global food system to make it healthier and more environmentally sustainable, mainly due to the world population projections that point to 10 billion people in 2050 (Willett et al., 2019).

An issue that must be highlighted is the loss of food along the supply chain. In addition to being expressive, reaching one-third of all food produced for human consumption, especially fruit and vegetables (Gustavsson et al., 2011), it also competes for limited natural resources (Lemaire and Limbourg, 2019). In this sense, food loss has a double impact on the environment: the excessive production of food and the management of food losses. In short, food loss is related to the waste of all resources used along the food chain, from agricultural production to consumption, including energy, human labor, and natural resources (Corrado et al., 2017).

Therefore, it is necessary to include these aspects to decipher the real environmental impact of food loss (FAO, 2014a, 2013).

It is estimated that the global loss of food and waste emits 4.4 GtCO<sub>2</sub>eq annually, considering changes in land use, corresponding to approximately 8% of the total global emissions of GHG (FAO, 2015b). In addition, it occupies 0.9 million hectares of soil and consumes 306 km<sup>3</sup> of water (FAO, 2014b).

Currently, several definitions of food loss and food waste are proposed. The FAO definition (2011) is widely accepted, used, and was adopted in this chapter, as stated above. However, waste resulting from inedible parts of foods or their processing as well as the loss and waste of edible parts of food along the food supply chain are also considered in these other definitions. Thus, in this work, we denominate as “residues” the processing waste residues, whether they are edible (e.g., fruit peel) or inedible (e.g., fruit seed), as well as the naturally inedible parts (e.g., some fruit leaves), based on Teigiserova et al. (2019b).

The food processing industry generates approximately 140 billion tons of residues annually. These residues are, in most cases, underutilized, even though they are rich in nutrients and bioactive compounds that could be used as bioproducts and/or raw material for products with higher added value (Hang, 2004; Zuin and Ramin, 2018).

Actions to reduce food loss and waste, as well as food residues, are necessary and encouraged, to contribute to a more sustainable and resilient food system (Lemaire and Limbourg, 2019; Willett et al., 2019) and guarantee food security (IPCC, 2019), as previously discussed. However, as it is difficult to eradicate residues in food production (Lemaire and Limbourg, 2019; IPCC, 2019), it is necessary to outline ways of using such residues that result in fewer environmental impacts than, for example, landfills, which are one of the main destinations of food residues (Ng et al., 2020; Papargyropoulou et al., 2014).

Currently, with the valorization of green industrial processes, the use of agro-industrial plant residues has been identified as an important strategy to develop sustainable products with greater added value for the chemical, pharmaceutical, and food industries (Freitas et al., 2021), given the rich and heterogeneous chemical composition of these residues (Fidelis et al., 2019; Jiménez-Moreno et al., 2020; Majerska et al., 2019) and their low cost (Zuin and Ramin, 2018). This is possible because food residues present a variable chemical composition on the basis of their generation and origin (Kavitha et al., 2020). They stand out for their high nutritional value due to the presence of satisfactory amounts of proteins, lipids, starch, micronutrients, bioactive

compounds, and dietary fibers (Banerjee et al., 2017; Faustino et al., 2019), providing numerous alternatives for the use by the food industry, which may improve the nutritional characteristics of food (Sancho et al., 2015). In addition, the use of this rich resource, mostly fruit and vegetables, could directly benefit local communities, through the formulation of new foods and the strengthening of existing ones (Lai et al., 2017). Many studies are using treated agro-industrial residues as ingredients for food products of greater nutritional value (Table 2).

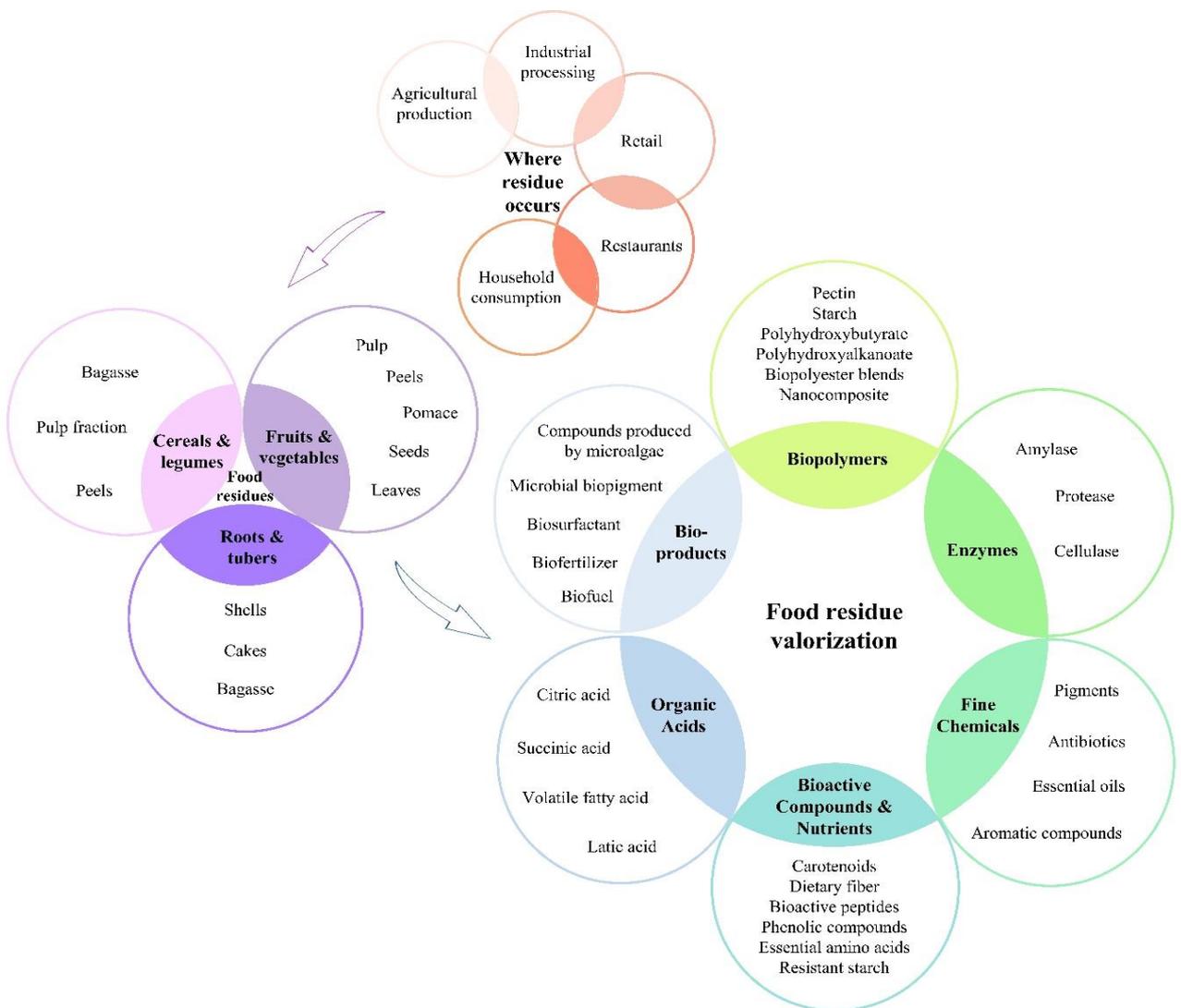
**Table 2.** Studies that used agro-industrial residues to improve the nutritional value of food products between 2020 and 2021.

<b>Agro-industrial residue</b>	<b>Compounds</b>	<b>Application</b>	<b>Reference</b>
Cocoa	Polyphenols	Chocolate/cocoa drinks	Manuela et al., 2020
Beet leaves extract	Phenolics and antioxidants	Fruit and vegetable smoothie	Fernandez et al., 2020
Peppermint hydrodistillation by-products	Phenolics	Ice creams	Berkas and Cam, 2020
Apple pomace	Dietary fibers and phenolics	Yogurt and yogurt drinks	Wang et al., 2020
Banana peel	Phenolics	Yogurt	Kabir et al., 2021
Apple peels and grape seeds	Phenolics and flavonoids	Yogurt	Brahmi et al., 2021
Carrot waste extract	$\beta$ - carotene	Yogurt	Šregelj et al., 2021
Apple pomace flour	Polyphenols	Yogurt	Jovanović et al., 2020
Mango and potato peel flour	Dietary fibers and polyphenols	Yogurt	Pérez-Chabela et al., 2021
Grape pomace powder	Polyphenols	“Primosale” cheese	Gaglio et al., 2021
Pomegranate peel	Phenolics	Cookies	Kaderides et al., 2020
Apple peel	Dietary fibers and polyphenols	Bread and wheat cookies	Nakov et al., 2020
Blackcurrant by-product	Dietary fibers and antioxidants	Chocolate cookies	Gagneten et al., 2021
Prickly pear peel flour	Dietary fibers and phenolics	Cookies	Bouazizi et al., 2020

Rice bran	Phenolics and antioxidants	Cookies	Christ-Ribeiro et al., 2021
Cashew waste	Dietary fibers	Cookies and flour	Araújo et al., 2021
Guava peels and cashew bagasse	Protein	Cereal bars	Muniz et al., 2020
Coffee silverskin	Dietary fibers, protein and polyphenols	Extruded cereal-based ready-to-eat food product	Beltrán-Medina et al., 2020
Unripe papaya powder	Dietary fibers and polyphenols	Pancake	Joymak et al., 2021
Flour and coconut residue	Dietary fibers	Pasta	Sykut-Domańska et al., 2020
Citrus reticulata (Kinnow) pomace	Dietary fibers and antioxidants	Pasta	Singla et al., 2020
Grape pomace	Phenolics	Pasta	Tolve et al., 2020
Onion skin powder	Dietary fibers, phenolics and flavonoids	Pasta	Michalak-Majewska et al., 2020
Grape and olive pomace	Dietary fibers and phenolics	Pasta	Balli et al., 2021
Whey and grape peels powders	Dietary fibers	Pasta	Ungureanu-Iuga et al., 2020
Olive pomace	Polyphenols	Bread and pasta	Cedola et al., 2020
Wheat bran	Protein and essential amino acids	Bread and pasta	Alzuwaid et al., 2021
Onion powder and onion peel extract	Dietary fibers and protein	Bread	Masood et al., 2020
Broad bean hull	Dietary fibers	Bread	Ni et al., 2020
Prickly Pear Peel Flour	Betalains and flavonoids	Bread	Parafati et al., 2020
Whole green banana flour	Dietary fibers and resistant starch	Bread	Khoozani et al., 2020
Milk permeate, Psyllium husk and apple by-products	Galactooligosaccharides (GOS) and lactic acid bacteria, desirable hydrocolloids and phenolic compounds	Nutraceutical chewing candy	Zokaityte et al., 2021
Sweet potato peel flour	Minerals, carbohydrate and dietary fiber	Beef hamburger	Marconato et al., 2020
Pumpkin peel flour	Minerals, carbohydrate and dietary fiber	Beef hamburger	Hartmann et al., 2020

Furthermore, these sources are considered good raw-materials since they can be bioconverted into high-value bioproducts, e.g., biofuel, bioactive compounds, enzymes, fine

chemicals, and biodegradable plastics (Ng et al., 2020; Sharma et al., 2021). Figure 5 presents the origin of food residues along the food supply chain, highlighting the main vegetable sources and types, as well as potential applications. Protein and lipid-rich food residues are suitable for animal feed, whereas cellulosic-rich food residues are suitable for cattle feed (Kavitha et al., 2020). Some residues may not be appropriate for the elaboration of technological and biotechnological products owing to their complexity, uncontrolled spoilage, and lack of traceability. In these cases, methods such as biogas production and composting can be applied (Poovazhahi and Thakur, 2020).



**Figure 5.** Food residues: Sources, types and opportunities for their valorization.

Residues and by-products from the industrial processing of fruit and vegetables are exploited for animal feed (Papargyropoulou et al., 2014; Velarde et al., 2020), biofuel manufacture (Abdelhady et al., 2020; Suhartini et al., 2020), recovery of bioactive compounds (Jiménez-Moreno et al., 2020; Rahimi and Mikani, 2019; Sánchez-Camargo et al., 2019) that can be applied as sustainable and potentially functional ingredients (Fidelis et al., 2020), enzyme production (Costa et al., 2017; Mojumdar and Deka, 2019), biofertilizers (Du et al., 2018; Lim and Matu, 2015), raw material for application in microalgae cultivation (de Medeiros et al., 2020; Koutra et al., 2018), pigment production (Arikan et al., 2020; Aruldass et al., 2016; Lopes and Ligabue-Braun, 2021), of biosurfactants (Jadhav et al., 2019; Rivera et al., 2019) and nanoparticles (Huang et al., 2020; Shruthy and Preetha, 2019), in addition to the extensive development of active and biodegradable packaging for food (Luchese et al., 2019; Vedove et al., 2021).

There are several studies in the literature that highlight the potential of extracting bioactive compounds from agro-industrial residues. Vodnar et al. (2017) identified bioactive compounds in some plant residues using different treatments and, in some of them, observed antioxidant, antimutagenic, and antimicrobial activities. Velarde et al. (2020) evaluated the content of phenolic compounds and antimicrobial potential of hydroalcoholic extracts from three plant residues – avocado, guava, and cherry plum leaves – and suggested their application in additives for animal feed. Sánchez-Camargo et al. (2019) recovered carotenoids from mango peel and observed their action against lipid oxidation in sunflower oil, acting as a natural antioxidant. Rahimi and Mikani (2019) recovered lycopene from tomato residues employing an edible solvent (sunflower oil) in a green process to generate a safe and high-quality extract. Fidelis et al. (2020) observed the effect of lyophilized camu-camu seed extract on the antioxidant and sensory characteristics of yogurt. The extract showed high phenolic content, in addition to high antioxidant activity. It was also able to inhibit the cell proliferation of two cancer cell lines and the in vitro activity of some enzymes of clinical importance. The addition of the extract to yogurt in different concentrations proved to be adequate from a sensory point of view and was able to increase the antioxidant and chemical reducing properties in yogurts.

Other application examples involve the production of enzymes. Mojumdar and Deka (2019) compared some agro-industrial residues as substrates to produce alpha-amylase through solid-state fermentation using *Bacillus amyloliquefaciens*. The substrates of wheat bran and potato skins, both alone and in combination, showed higher enzymatic production when

compared to rice bran. Moreover, the purest and most active amylases were those obtained with wheat bran substrate alone and combined with potato skins.

Another possibility is the production of biofertilizers from vegetable residues, which was explored by Lim and Matu (2015) using solid-state fermentation. Biofertilizers made from watermelon, papaya, and banana residues proved to be suitable for plant treatment; watermelon resulted in the greatest weight and the average length from mustard plant samples.

The use of agro-industrial residues as a means to produce biosurfactants is also extensively explored. Jadhav et al. (2019) produced a sophorolipid based on sunflower oil acid by submerged fermentation using *Starmerella bombicola*, which showed better emulsification and wetting properties and less foaming than the polysorbate 20 synthetic surfactant evaluated. Paraszkiwicz et al. (2018) identified and characterized lipopeptide biosurfactants produced by two strains of *Bacillus subtilis* grown in various media prepared from plant residues, including extract from apple and carrot peel. Studies like these reinforce the potential for the development of biosurfactants using residues as a fermentative medium, since they are widely generated and low in cost, and can therefore contribute to better managing these residues.

In addition, the residues can also serve as an alternative culture medium for the development of microalgae. De Medeiros et al. (2020) evaluated the use of agro-industrial residues from fruits and vegetables as an alternative means for the cultivation of three freshwater microalgae, resulting in adequate cell growth, absence of chemical and microbiological contaminants, in addition to better antioxidant activity and mono and polyunsaturated fatty acid levels in comparison with the conventional synthetic medium. The use of this type of lower-cost raw material can improve the cost-benefit ratio of microalgae cultivation and contribute to the production of compounds of interest to the food industry as well as other products with ecologically correct applications (Koutra et al., 2018).

Some biopigments have already been produced by microorganisms using agro-industrial residues as raw material. Arikan et al. (2020) used by-products by processing apple, pomegranate, black carrot, and red beet as a substrate for the filamentous fungus *Aspergillus carbonarius* to produce pigments. The authors observed the great potential of pomegranate pulps in pigment production, especially yellow, suggesting that *A. carbonarius* can produce melanin. Aruldass et al. (2016) optimized the production of yellowish-orange pigment by cultivating *Chryseobacterium artocarp* CECT 8497 in a pineapple residue liquid medium. In this work, the researchers were able to produce a bacterial pigment, on a pilot scale, using a

cheaper and more economical medium than the usual nutrient broth. These works highlight the potential of using vegetable residues as an alternative growth medium to produce biopigments.

Several studies have evaluated the incorporation of agro-industrial residues, such as blueberry peel and powdered jaboticaba (Luchese et al., 2019) and grape skins (Vedove et al., 2021), in biodegradable starch-based films, as a potential colorimetric indicator for application as smart food packaging. In these studies, the potential of the anthocyanins in the residues to change the color of the films in response to changes in the pH of the medium was observed. Agro-industrial residues are also used for the elaboration of nanoparticles. Shruthy and Preetha (2019) developed cellulose nanoparticles prepared from potato skins through alkaline treatment, bleaching, and acid hydrolysis, and incorporated them in an ecological film based on biodegradable polyvinyl alcohol added to fennel seed oil. The nanoparticles developed provided greater tensile strength to the films, in addition to good transparency, thermal stability, and biodegradability. Huang et al. (2020) developed modified cellulose nanofibrils from cassava residue as a reinforcing agent for starch-based films, resulting in a biodegradable film with hydrophobic characteristics and good mechanical and barrier properties.

Another interesting possibility is the use of fruit that would otherwise be discarded due to difficulties in their commercialization, as raw materials for the elaboration of biodegradable films, thus contributing to reducing losses (Matheus et al., 2021, 2020b). For example, persimmon, which has high rates of waste in Brazil (Matheus et al., 2020a), showed great potential regarding the development of biodegradable films used as packaging for minimally processed vegetables (Matheus et al., 2021).

Finally, despite several studies addressing some options for the use of agro-industrial residue, it is necessary to evaluate cases on an individual basis to understand the real environmental impacts of alternative uses, to guarantee that they cause less degradation to the ecosystems and are inserted in the context of the circular economy and sustainable development.

#### **4. CIRCULAR ECONOMY: A HOLISTIC APPROACH**

It is already evident that food loss is a global problem (Corrado et al., 2017; Laso et al., 2021) and still generates large amounts of agro-industrial residues. Food loss, food waste and food residues are often poorly managed, culminating in negative impacts on different sectors of human life and the environment (Papargyropoulou et al., 2014). Thus, it is essential to search

for solutions that consider socioeconomic, nutritional, and environmental aspects. In this sense, one possible approach is a circular economy, which can contribute to human activities that favor the reduction of waste generation by transforming such waste into new resources to be exploited, making it possible to align economic development with the promotion of environmental quality and social equity under the spectrum of human rights (Velenturf and Purnell, 2021). From this perspective, the residues generated in the food industry can go from being merely waste to becoming renewable resources with potential applications in different sectors.

In this context, the SDGs stand out as a strategic, global project of sustainable development linked to the circular economy (Schroeder et al., 2019). Their main purposes are to eradicate poverty, reduce inequality and minimize climate change, preserving oceans and forests (UN, 2015). The use of agro-industrial residues could contribute directly or indirectly to some SDG targets (Figure 6) since this strategy integrates environmental, socioeconomic, and nutritional issues.



**Figure 6.** The UN’s 17 Sustainable Development Goals highlighting those that may be favored when using food residues (2, 7, 8, 11, 12, 14, and 15).

Looking at it in greater detail, using food residues can minimize the environmental impact, thus contributing to goals 11, 12, 14, and 15, more specifically, subitems 11.4, 11.6, 14.1, and 15.5. More environmentally conscious management allocates the residues to non-polluting purposes, in order to assist in substituting non-renewable resources for the production

of energy, bioplastics, and chemicals (Freitas et al., 2021). Therefore, harnessing food waste and food residues can contribute to the protection of the world's natural heritage and a reduction in the environmental impact of cities (11.4 and 11.6), a lower release of waste into oceans and coastal regions (14.1), and protection of biodiversity as a result of less environmental degradation (15.5) (UN, 2015).

The use of waste and residues is directly related to goal 12, since it addresses the need to change current consumption and production patterns to a more sustainable model. In general, it is expected: to use and manage natural and renewable resources more efficiently, in addition to adopting actions towards the gradual transition to the circular economy model (12.1 and 12.2); halve world food waste per capita (12.3); reduce the generation of waste based on different strategies (prevention, reduction, recycling, and reuse) (12.5); and to properly manage chemical products and residues, avoiding contamination of different ecosystems and impacts on the environment and human health (12.4) (UN, 2015).

Given the wide range of compounds, versatility, and potential of agro-industrial plant residues (Jiménez-Moreno et al., 2020), their by-products can be extensively explored to contribute indirectly to the different SDG subitems. For example, the development of nutritionally richer ingredients and/or food products may favor a healthier diet (Majerska et al., 2019), especially for poor and vulnerable people, thus relating to subitem 2.2 which aims to end all forms of malnutrition. If part of the waste is destined to produce biodegradable food packaging (Moura et al., 2017), capable of replacing conventional plastic packaging of petrochemical origin, environmental pollution, especially in the oceans, can be minimized. In addition, taking advantage of liquid residues from food processing can contribute to such residues not being disposed of in rivers, which leads to a series of ecological imbalances. One example is the industrial effluent from cassava processing that can be used to produce biogas and biosurfactants or as a growth medium for microorganisms (Zevallos et al., 2018). These applications can contribute to the prevention and reduction of marine and coastal pollution (14.1). If the use of waste is destined to produce fuel and renewable energy (Suhartini et al., 2020), thus relating to subitem 7.2, as it aims to increase the global percentage of renewable energy. Finally, the increase in the efficiency of global resources in both consumption and production, will contribute to subitem 8.4, making it possible to combine economic growth and sustainability from the circular economy.

Despite the potential to apply agro-industrial residues as more sustainable alternatives, it is essential to carry out scientific research that analyzes specific cases of exploitation from the point of view of the circular economy, assessing whether the environmental, economic, and social impacts are indeed positive. Velenturf and Purnell (2021) carried out an extensive analysis of the circular economy, demonstrating that, in many cases, theory and practice have been contradictory, either not being truly in line with what is proposed or even negatively impacting the environment and society. They stressed that sustainable development can be achieved with the support of circular economy precepts, but that it is necessary to take a more holistic approach in understanding these concepts, so that it is not reduced to closed-loop recycling with short-term economic benefits (Velenturf and Purnell, 2021).

It is also important to guarantee that these strategies do not stimulate the generation of residues, contradicting one of the most important modern-day needs: to prevent food loss and the generation of waste (Laso et al., 2021; Teigiserova et al., 2019a). Therefore, coordinated actions between all the actors involved (society, governments, and industries) must be strengthened to create a culture of circular economy and sustainable development, guaranteeing economic growth, environmental quality, well-being, and social equity for current and future generations (Velenturf and Purnell, 2021).

## **5. FUTURES PERSPECTIVES**

The use of agro-industrial residue can increase economic gains, reduce production costs and increase the value of residues, increase the nutritional value of food, in addition to strengthening sustainable practices inserted in the circular economy. These changes are in line with reducing our anthropogenic footprint on an already extensively affected planet (Nazzaro et al., 2018; Willett et al., 2019).

Due to the diverse and interesting chemical composition, agro-industrial plant residues can be better utilized from a circular economy perspective, in which bioconversion can generate different bioproducts with potential socioeconomic benefits coupled with lower environmental impact (Fidelis et al., 2019; Freitas et al., 2021; Kavitha et al., 2020; Ng et al., 2020).

The need for changes in the food sector and waste management is increasingly urgent. The agreed deadlines for achieving the SDG targets are approaching and there are still many adjustments to be made. Furthermore, with the global crisis generated by the COVID-19 pandemic, it will be necessary to devise strategies for a worldwide better recovery. Efforts must

be made to mitigate negative impacts and prevent them from lasting for many years. It is essential to make the food system more resilient, reducing food losses and waste generation, as well as environmental, socioeconomic, and nutritional setbacks.

Food waste management strategies must consider the numerous challenges involved to achieve optimal performance in the environmental, health, economic and social dimensions. As food waste is heterogeneous, knowing its composition is one of the determining factors for the most appropriate treatment approach, as well as the necessary infrastructure to be developed in each region in order to properly proceed with the collection, segregation, and processing of different food waste (Arumdani et al. 2021; Thakur et al., 2020; Singh, 2020).

There are several processes for treating food waste, such as biological, physical, and chemical methods. Currently, the most common waste destination is sanitary landfills, representing the destination of 80% of solid waste discarded (Kaushik and Sharma, 2020), followed by incineration, which causes environmental consequences (Thakur et al., 2020). Arumdani et al. (2021) observed that among the five largest countries that generate waste in Southeast Asia, only Indonesia and Thailand classify and recycle municipal solid waste, which is 64% organic or food waste.

Further advances must also be made towards efficiently reducing food loss by improving data on the quantification, causes, and destination of food waste (Hartikainen et al., 2020). In this sense, Vandana et al. (2020) point out that integrated and technological systems can contribute to better management of the entire food supply chain, reducing waste and sustainably supporting the population. Finally, expanding the techno-scientific use in the stages of post-harvest, transport, storage, and food processing can contribute to increasing the shelf life of food, preserving and/or increasing its sensory and nutritional quality. In the future, more actions related to sustainable development, linked to the circular economy, are expected to be taken. Therefore, initiatives for the reuse of food waste are of great importance and interest and must be encouraged by all social actors.

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## CAPÍTULO II

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**Biopolymers as green-based food packaging materials: a focus on modified and unmodified starch-based films**

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Title Short Version: Starch-based food packaging films

## **ABSTRACT**

The ideal food packaging materials are recyclable, biodegradable, and compostable. Starch from plant sources, such as tubers, legumes, cereals, and agro-industrial plant residues, is considered one of the most suitable biopolymers for producing biodegradable films due to its natural abundance and low cost. The chemical modification of starch makes it possible to produce films with better technological properties by changing the functional groups into starch. Using biopolymers extracted from agro-industrial waste can add value to a raw material that would otherwise be discarded. The recent COVID-19 pandemic has driven a rise in demand for single-use plastics, intensifying pressure on this already out-of-control issue. This review provides an overview of biopolymers, with a particular focus on starch, to develop sustainable materials for food packaging. This study summarizes the methods and provides a potential approach to starch modification for improving the mechanical and barrier properties of starch-based films. This review also updates some trends pointed out by the food packaging sector in the last years, considering the impacts of the COVID-19 pandemic. Perspectives to achieve more sustainable food packaging toward a more circular economy are drawn.

**Keywords:** Agro-industrial waste; Biodegradable films; Starch-based films; SARS-Cov2 pandemic; Long-term impacts; Sustainable Development Goals.

## 1. Introduction

The demand for plastics has been growing all over the world since the 1950s (Figure 1) (Geyer, Jambeck, and Law 2017). Average global production of plastic stands at approximately 400 million tons and is expected a growing around 600 million tons in 2025. About 1/3 of production is destined for the packaging sector, mostly single use (PLASTICSEUROPE 2020; Arkin et al. 2019; UNEP 2018). These synthetic plastics have been contributed to the global pollution including the both terrestrial and aquatic ecosystems, accumulating microplastics in the oceans, and marine animals and to higher emissions of greenhouse gases (Asgher et al. 2020; Geyer, Jambeck, and Law 2017; Boucher and Billard 2019). Thus, the high disposal of plastics impacts negatively the biosphere and contributes to climate change, as well as being one of the most damaging human activities in terms of environmental changes that characterize the current geological era known as the Anthropocene (Acquavia et al. 2021; Willett et al. 2019).

However, plastic has been essential for, virtually all industrial sectors, such as packaging. The use of food packaging is primarily intended to protect food from the surrounding environment, maintaining its quality, safety and extending its shelf life, and in turn helping to reduce food losses and wastage (Rojas et al. 2019; J. W. Han et al. 2018). Besides, the packaging facilitates the handling in the transport and storage of food, provides important information to the consumer, and promotes and encourages the sale of the food product (Rodríguez-Rojas et al. 2019; Yildirim et al. 2018). In addition to these basic functions, other requirements in food packaging such as biodegradability and bioactivity (J. W. Han et al. 2018; Santos et al. 2021), have become necessary over the years as discussions on consumer health and environmental impacts of different materials have intensified (Rojas et al. 2019; J. W. Han et al. 2018).

Currently, efforts are taking place to expand the use of alternative materials made from synthetic, to meet sustainability and food safety guidelines (Rojas et al. 2019). The urgency to modernize food packaging materials has been reinforced by the increased use of plastics during the coronavirus (COVID-19) pandemic (Parashar and Hait 2021; A. L. P. Silva et al. 2021; Barone et al., 2021), given the increased health concerns and consumer awareness of the importance of packaging for food safety (Barone et al. 2021; Vanapalli et al. 2021; Southey 2020). The projected compound annual growth rate of the global packaging market during the pandemic is around 6%, with the plastic packaging segment leading the market (Markets

Business Insider 2020). The future of food packaging materials envisages recyclable, biodegradable, or compostable materials (J. W. Han et al. 2018; Zhong et al. 2020), as well as being non-toxic to the environment and humans (J. W. Han et al. 2018). In addition, future trends envisage packages that intentionally and positively interact with food, providing greater nutritional, sensory, and hygienic quality, extending the shelf life of packaged foods (active packaging), and/or monitoring food quality and safety conditions based on stimuli responses (intelligent packaging) (J. W. Han et al. 2018; Yildirim et al. 2018; Almasi, Oskouie, and Saleh 2020; Bhardwaj, Alam, and Talwar 2019; Valencia, Luciano, and Fritz 2019). Active packaging is basically composed of a polymeric matrix, that is a barrier, and active compounds (active layer), which can interact by different mechanisms in a wide range of food matrices (Pascuta and Vodnar, 2022; Azman et al. 2022). Regarding intelligent packaging systems, compounds can be incorporated into the polymeric matrix as indicators, data carriers, sensors, and other intelligent systems, in order to monitor the parameters of food quality (Azman et al. 2022).

In this context, agro-industrial residues have been widely investigated as possible precursors of materials for food packaging that meet these current demands (Santos et al. 2021; Acquavia et al. 2021; Barone et al. 2021; Matheus et al. 2021), as they are rich in macromolecules and bioactive and aromatic compounds (Valencia et al. 2021), in addition to being often biodegradable, biocompatible, and non-toxic (Yildirim et al. 2018; Maraveas 2020). Biopolymers based on proteins and carbohydrates are some of the most studied in terms of the production of food packaging (Asgher et al. 2020), with emphasis on macromolecules extracted from fruit and vegetable waste (Acquavia et al. 2021) (Figure 2). In line with this context, Luchese et al. (2021) recently developed cassava starch-based films incorporated with orange residue from the juice industry. Among the main results, the authors observed that the residues (hull and bagasse), given the richness of lignocellulosic compounds, were able to increase the stiffness and reduce the flexibility of the starch film. However, it is important to emphasize that the fibers as a reinforcing agent in the film was evidenced only when the residues were used in the form of powder, rather than an aqueous extract. When the film was added to aqueous extract it exhibited greater water solubility and greater flexibility (Luchese et al. 2021). Otoni et al. (2018) developed an optimized film formulation based on carrot residues and hydroxypropylmethylcellulose with suitable properties for food packaging application, namely 30 MPa tensile strength (TS), 3% elongation at break (EB), 2 GPa Young's modulus (YM). In addition, the authors observed a high rate of aerobic biodegradation of the optimized film in the

soil ( $\pm 40$  mL accumulated CO<sub>2</sub>) during 75 days of analysis, in addition to the possibility to expand the production scale, which is, however, subject to changes in mechanical performance (Otoni et al. 2018). Martelli-Tosi et al. (2018) studied the influence of the addition of nanocelluloses produced from soybean straw as reinforcing agents in soy protein isolate films. Both nanocelluloses obtained by acidic and enzymatic hydrolyses increased the TS of the film by 38 and 48%, respectively, compared to the control film (without nanocelluloses in the film formulation). The authors concluded that the nanocellulose obtained by enzymatic hydrolysis showed adequate incorporation in the protein matrix, favoring the mechanical and water vapor barrier properties in the soy protein film (Martelli-Tosi et al. 2018). These works corroborate the potential use of agro-industrial residues as a source of biopolymers for the development of innovative food packaging materials.

In addition to these prominent biopolymers, other molecules are promising sources of biodegradable films, such as gelatin, collagen, wheat gluten, chitin/chitosan, and pectin (Figure 2) (Zubair and Ullah 2020; Hassan et al. 2018). Starch is considered one of the most relevant biopolymers to produce biodegradable films, and replace conventional plastics, given its natural abundance, good cost-benefit, and significant biodegradation capacity (Zhong et al. 2020; Asgher et al. 2020; Yang, Ching, and Chuah 2019). Different sources of starch extraction have been investigated for film production, such as cassava (Luchese et al. 2021), corn (Luchese et al. 2019), potatoes (La Fuente et al. 2020), and fruit (Costa et al. 2021; A. P. M. Silva et al. 2019; Tirado-Gallegos et al. 2018). It is important to highlight that the use of biopolymers extracted from agro-industrial by-products in new materials strengthens the circular economy precepts, contributes to the reuse and valorization of commonly underused raw materials, and minimizes the environmental impacts associated with the management of agro-industrial waste (Barone et al. 2021; Zhong et al. 2020). In addition, the use of biopolymers from agro-industrial wastes is aligned to some of the Sustainable Development Goals, such as 12.3 and 12.5 that aim, respectively, to halve world food waste *per capita* by 2030 and reduce the generation of waste based on different strategies (prevention, reduction, recycling, and reuse) (UN 2015).

Therefore, a deeper understanding of the role of biopolymers as materials for food packaging could lead to the improvement and expansion of this sector within the food industry, in addition to environmental and health benefits for consumers. This review provides an overview of the use of biopolymers, with a particular focus on starch, to develop sustainable materials for food packaging. This review provides an overview of biopolymers, with a

particular focus on starch, to develop sustainable materials for food packaging. This study summarizes the methods and provides a potential approach to starch modification for improving the mechanical and barrier properties of starch-based films. In addition, this review highlighted some trends for the food packaging sector, considering the impacts of the COVID-19 pandemic and the perspective to apply more sustainable materials.

## **2. New green materials for food packaging development**

Environmental concerns coupled with changing consumer preferences are fueling the growing search for new, more sustainable food packaging materials in the food industry (Figure 1) (J. W. Han et al. 2018; Asgher et al. 2020). In 2020, approximately 1.2 million tons of biodegradable bioplastics were produced, corresponding to 60% of the total production of bioplastics. The most representative were starch blends and polylactic acid (which individually represented 18.7% of the total bioplastics produced). In addition, it is estimated that, in 2025, the production of biodegradable bioplastics will stand at around 1.8 million tons. The packaging sector is the largest field of application of bioplastics, corresponding to approximately 0.99 million tons (47%) of the total bioplastics market in 2020 (European Bioplastic 2020). In this context, the use of biopolymers in the food packaging sector is promising, since they are generally renewable, biodegradable, non-toxic, abundant, and economical (Asgher et al. 2020; Yildirim et al. 2018). Biopolymers can be obtained through chemical/biotechnological processes (such as polylactic acid and polyhydroxyalkanoates) or extracted from biomass (such as polysaccharides and protein) (Asgher et al. 2020; Arrieta et al. 2017). Some of the most studied biodegradable biopolymers for food packaging applications are gelatin (Said, Howell, and Sarbon 2021), collagen (Valencia et al. 2019), wheat gluten (Alonso-González et al. 2021), soy protein (Y. Han, Yu, and Wang 2018), chitin/ chitosan (Priyadarshi and Rhim 2020), lignocellulosic compounds (Chong, Law, and Chan 2021), pectin (Mellinas et al. 2020), and starch (Lauer and Smith 2020). Table 1 presents some recent studies on the development of biopolymer-based packaging materials.

Proteins, both of animal and vegetable origin, can form resistant, elastic biodegradable films (a strong and cohesive polymeric network), with low permeability to oxygen and aromas, which favors their application as food packaging. However, the filmogenic capacity of proteins is influenced by the source and its molecular characteristics, which also impacts the mechanical properties of the films (Calva-Estrada, Jiménez-Fernández, and Lugo-Cervantes 2019). For

example, collagen fibers have a lower film-forming capacity compared to collagen in powder form, making their use for the development of biodegradable films difficult (Valencia et al. 2019; Ma et al. 2020). In contrast, wheat gluten is considered a protein with a high capacity to form biodegradable films (Zubair and Ullah 2020), with selective barrier properties to gases, like oxygen, carbon dioxide, and aromatic compounds, and viscoelastic heat-sealing properties. The disadvantages, however, are that wheat gluten-based films have high moisture sensitivity and water vapor permeability (WVP), in addition to a relatively fragile structure, which makes their use as food packaging challenging (Zubair and Ullah 2020; Calva-Estrada, Jiménez-Fernández, and Lugo-Cervantes 2019). Calva-Estrada, Jiménez-Fernández, and Lugo-Cervantes (2019) state that protein films with superior characteristics are those based on gelatin, myofibrillar proteins, whey protein, wheat gluten, soy protein, and corn zein. Zubair and Ullah (2020) report that whey, soy, and wheat gluten protein-based biodegradable films are the most studied materials in terms of food packaging. In addition, plasticization, crosslinking techniques, and association with other polymers and nanocompounds are some of the improvement strategies being constantly developed and employed for protein-based films in order to make them more competitive with plastics of petrochemical origin (Calva-Estrada, Jiménez-Fernández, and Lugo-Cervantes 2019; Ma et al. 2020; Valencia et al. 2019).

Carbohydrate-based polymers have been widely explored as films or coatings for food packaging (Zubair and Ullah 2020). Regarding the use of agro-industrial residues to produce biodegradable films, in general, starch and lignocellulosic compounds are extracted from fruit and vegetable residues (Acquavia et al. 2021). Lignocellulosic compounds are natural fibers composed of cellulose (35–50%), hemicellulose (20–35%), and lignin (10–25%), the composition of which varies according to the plant origin, be it from wood or non-wood fibers (Chong, Law, and Chan 2021). Cellulose-based films have good mechanical resistance, high surface gloss, and transparency (Asgher et al. 2020). The higher the cellulose content in the filmogenic formulation, the more resistant the films are, which also negatively affects their biodegradability (Maraveas 2020). Hemicellulose-based films commonly exhibit low oxygen permeability and are more brittle, which increases the need to incorporate plasticizers to increase their flexibility and applicability as food packaging (Asgher et al. 2020). Lignin, a polyphenolic macromolecule, improves the mechanical resistance of films and provides antioxidant properties, favoring the development of a package that is not only biodegradable but possibly active (Yang, Ching, and Chuah 2019).

Starch, the most abundant polysaccharide in nature, is considered an important carbon precursor for the production of biologically-based polymers and can be obtained from different plant sources, such as tubers, legumes, cereals, and agro-industrial wastes, especially by traditional crops (Agarwal 2021; Maraveas 2020). In addition, starch is renewable, biodegradable, low cost, Generally Recognized As Safe (GRAS) to package food products. It shows structural and crystallinity characteristics that favor its transformation into a thermoplastic material with adequate properties for film production (Agarwal 2021). Differently that animal-based biopolymers, starch can be used as a packaging precursor for plant-based and vegan food products – an increasing trend in many countries. Su et al. (2022) reinforced the main aspects that make starch films (isolated or blended) an emerging material for food packaging are simple production, desirable functional properties, high rate of degradability after use and low digestibility.

Dai, Zhang, and Cheng (2019) compared starches extracted from various sources, namely six native starches (cassava, waxy corn, potato, sweet potato, corn, and wheat) and three commercial modified cassava starches. Films based on modified cassava starch, especially cross-linked starch, showed greater strength (3.0–10.1 MPa of TS), flexibility (66.4–181.1% of EB) and barrier property ( $\pm 1.1$ – $1.5 \times 10^{-11} \text{ g m}^{-1} \text{ s}^{-1} \text{ Pa}^{-1}$  of WVP) when compared to films based on native starch. The potato starch-based film had better mechanical properties and the cassava and sweet potato starch-based films had a higher barrier property compared to native starches. The authors suggested that the amylose content and the structural modifications in starch directly interfere in the physicochemical properties of the films (Dai, Zhang, and Cheng 2019).

Similarly, Żołek-Tryznowska and Kałuża (2021) compared starches extracted from maize, potato, oat, rice, and tapioca, regarding their film-forming capacity and their properties. The study observed that the potato starch film was more resistant (3.05 MPa of TS) compared to the oat starch film (0.36 MPa of TS). Concerning wettability and hydrophilicity, the lowest values found were for oat and tapioca films. The authors concluded that despite the disadvantages found in starch films compared to other materials commonly used as biodegradable packaging, improvements in these starch-based films should still be investigated, especially for use as disposable packaging (Żołek-Tryznowska and Kałuża 2021). It is clear that the different biopolymers will form materials with distinct and unique properties (Maraveas 2020). It is noteworthy that changes in the chemical structure of these biopolymers can occur

and/or reinforcing agents, binders, and plasticizers can be added to filmogenic formulations, to improve the films' properties, without compromising biodegradability. This all takes place while favoring the application of this material as a more sustainable food packaging that guarantees the quality and extension of the food's shelf life.

Using biopolymers extracted from agro-industrial waste can add value to a raw material that would otherwise be discarded and contribute to reducing food losses and minimizing environmental impacts when compared to other more common waste management destinations, such as landfills and incineration (Ng et al. 2020). J. W. Han et al. (2018) signalizes that future projections point to continuous innovations in the packaging sector, as well as the development of new technologies so that food packaging materials are more sustainable. Given the importance of starch in the area of food packaging, this study will focus on the main aspects of using starch for the development of materials for food packaging and its influence on mechanical, barrier, and thermal properties.

### **3. Starch-based films: a promising material for food packaging**

Starch is one of the most abundant biopolymers in nature and is considered the main storage carbohydrate of plants. Starches are mainly composed of two distinct macromolecules, amylose and amylopectin (Figure 3), both of which contain  $\alpha$ -D- units (glucose) and break down into carbon dioxide and water (Susmitha et al. 2021; Bello Perez and Agama-Acevedo 2017). Amylose consists of  $\alpha$ -(1-4)-linked D-glucose, whereas amylopectin has the same backbone as amylose but with myriad  $\alpha$ -(1-6)-linked branch points. It can be used as raw material in foods and pharmaceutical products, fermentation, paperboard, adhesives, paints, and plastics. Commercial starches are obtained mainly from potato, wheat, maize, cassava, and rice (Bello Perez and Agama-Acevedo 2017; Zhang, Rempel, and McLaren 2014).

The ratio of amylopectin to amylose varies significantly depending on the source of the starch. The ratio of the two polysaccharides ranges from 70 to 82% amylopectin and 18 to 30% amylose for the most common cereal starches. However, some starches have a high amylopectin content (98–99%), known as *waxy* starches, and others have high amylose content (50–70%) (Bello Perez and Agama-Acevedo 2017; Buléon et al. 1998). For example, wheat, corn, and potato starches contain about 20–30% amylose, while waxy starches contain less than 5%. The amylose content in starches with high amounts can be as high as 50–80% (Zhang, Rempel, and McLaren 2014). These characteristics, as well as other aspects of starch granules,

such as shape and size, amylose/amylopectin ratio, chain length distribution and arrangement of components influence the functionality of this biopolymer (Bello Perez and Agama-Acevedo 2017; Buléon et al. 1998).

The behavior of the polysaccharide in solution can be correlated with some chemical-physical properties such as molecular weight, rotation, and hydrodynamic radius. The analysis of these molecular characteristics of the starch components is essential to produce derivatives, modifying its structure to develop new products or to extend its application. Mild conditions (pH, temperature, and process time) are necessary to maintain the starch structure intact when the method used to study its characteristics requires solubilization (Bello Perez and Agama-Acevedo 2017). Due to its lower relative decomposition temperature ( $\sim 230$  °C), when compared to its glass transition ( $T_g$ ) and melting ( $T_m$ ) temperatures, starch cannot be easily transformed into a material in its native form (Buléon et al. 1998). However, with the addition of plasticizers, heat, high pressure, and mechanical shear force during starch processing, a thermoplastic starch (TPS) can be obtained. Plasticizers disrupt hydrogen bonds, thus reducing intermolecular forces. Consequently, the flexibility of the starch polymer chains is increased and both  $T_g$  and  $T_m$  will be below their decomposition temperature, thus exhibiting a plastic material behavior (Narayan 2006; Samsudin and Hani 2017).

The amylose and amylopectin are packed together within the semicrystalline granule with an overall crystallinity of about 15–45% depending on its origin, and on the techniques used to determine it. To generate TPS that can be processed as conventional polymers, this complex semicrystalline structure needs to be “deconstructed” to produce an amorphous material (Ortega-Toro et al. 2017; Chaléat, Halley, and Truss 2014). Thermoplastic starch can be molded into different shapes or blown into films. Properties such as TS, EB, elastic modulus (EM),  $T_g$ , and gas barrier, are significantly affected by processing parameters, plasticizer content, moisture content, among others (Zhang, Rempel, and McLaren 2014). The starch source also influences the properties of TPS, and these differences in properties have been associated in part with differences in amylose and amylopectin content and their molecular structure (Buléon et al. 1998; Chaléat, Halley, and Truss 2014; Borges et al. 2015).

Mechanical properties stand out between the parameters of packaging polymer films since the films need to be strong enough to resist high external forces and provide perfect protection for inner articles (Hu, Chen, and Gao 2009). López and García (2012) reported that cassava and corn starches, containing 15.5% and 23.9% amylose, respectively, showed good

film-forming ability. The film made from corn starch, which contained more amylose, was more moisture resistant and less flexible than the cassava starch film (López and García 2012). The cassava starch, normal rice, and waxy rice-based films were evaluated and it was found that the films made from cassava starch were stronger and more water resistant than the others, due to the high amylose content (Zhang, Rempel, and McLaren 2014; Phan et al. 2005). Glycerol and sorbitol were used as plasticizer to produce rice, cassava, and potato starch films. The rice starch films produced using glycerol as plasticizer presented lower TS values than those produced with sorbitol, and EB values at over 100 times higher. Higher values of TS and lower EB were also found in films from potato and cassava starch when prepared with glycerol (Borges et al. 2015). Cassava starch films added with cinnamon essential oil were evaluated in structural, physical, mechanical, and thermal changes. It was verified a significant increase in the cinnamon essential oil content, which significantly increased the EB and improved the ductility of the film (Zhou et al. 2021). Potato, corn, wheat, and waxy corn starches plasticized with glycerol and water, showed a relatively high EB, low TS, and low YM, typical of a rubbery state. But the TPSs from different sources were clearly different: TPS with around 25% amylose (wheat, corn, and potato starches) showed higher TS than TPS with 99% amylopectin (waxy corn starch) (Chaléat, Halley, and Truss 2014). The study carried out by Żółek-Tryznowska and Kałuża (2021) evaluated films made from oat and tapioca starches. Those films showed lower TS (0.36 and 0.78 MPa, respectively), compared to potato starch films. Oposely, potato starch-based films showed the highest TS (3.05 MPa), albeit at levels 10 times lower compared to those for modern biodegradable packaging films (Żółek-Tryznowska and Kałuża 2021).

Low mechanical strength is one of the noteworthy shortcomings concerning starch-based polymers. Recent research has focused on overcoming these problems by, for example, blending starch with other polymers, choosing different kinds of plasticizer, and using starch sources with high amylose content (Zhang, Rempel, and McLaren 2014). Therefore, considering inherent drawbacks in starch-based films, eco-friendly, low-cost treatments can significantly improve their competitiveness, when compared to conventional plastics, in particular, for food packaging. These reinforcements can be compensate as starch has the advantages of being abundant, low-cost, biodegradable, compostability, non-toxicity, and edible, making it a promising alternative as a substitute for petroleum-based polymers (Żółek-Tryznowska and Kałuża 2021; Basiak, Lenart, and Debeaufort 2018). In addition, starch-based films and compounds offer great potential as ecologically suitable materials for food packaging

and the incorporation of active compounds can create packaging with antibacterial and antioxidant properties (Żołek-Tryznowska and Kałuża 2021; Chollakup et al. 2020). Cheng et al. (2019) developed starch film incorporated with eugenol with antibacterial activity, especially against *E. coli*, which showed good application as active packaging capable of extending the shelf life of pork. Starch-based films also have the potential to be the polymer base for intelligent food packaging. For example, Vedove et al. (2021) developed an intelligent packaging from cassava starch and anthocyanin, which acted as the natural pH indicator change in meat stored at 6 °C. Costa et al. (2021) also incorporated anthocyanin as a pH indicator in film of jackfruit seed starch. The authors observed the potential as an intelligent packaging to indicate the freshness of fish, presenting a perceptible color variation in the films within 48 hours (Costa et al. 2021). It is expected that future research on starch films will further explore this trend of active and intelligent packaging, even seeking to expand the use of agro-industrial residues for the extraction of bioactive compounds (Barone et al. 2021).

#### **4. Starch modification methods: the most common treatments to enhance starch-based films**

Starch-based films can be produced from different sources such as corn, cassava, potato, rice, ginger, and wheat (Alcázar-Alay and Meireles 2015; La Fuente et al. 2020). Although chemically similar, the starch sources, significantly, affect the film properties (Luchese et al. 2018b; Dai, Zhang, and Cheng 2019; Lauer and Smith 2020). The amylose:amylopectin ratio is the main factor that impacts the physical properties of starch films, since the higher the amylose:amylopectin ratio, the more rigid and crystalline the films. In addition, low amylose:amylopectin ratio hampers the production of films (Luchese et al. 2018b; Cano et al. 2014; Colussi et al. 2017; Lauer and Smith 2020). It is worth noting that factors such as the starch molecular weight, protein, plasticizer, and phosphate monoester content; and granule size can also affect these films (Lauer and Smith 2020).

Regarding the application as food packaging, generally speaking, the starch-based films must present enhanced properties in terms of texture, viscosity, adhesion, homogeneity, hydrophobicity, antimicrobial, and rigidity (Rhim, Park, and Ha 2013; Jaiswal, Shankar, and Rhim 2019). Moreover, starch-based films should present high TS and elasticity, as well as reasonable EB to provide robustness and resistance when exposed to various stresses during processing and shipping (Emblem and Emblem 2012).

In this sense, native starched-based films usually have limited properties (Tanwar et al. 2021). One strategy used to overcome these drawbacks includes the starch modification treatments such as ozone, gamma irradiation, plasma, ultrasound, dry heating treatment, and a combination of them (Lauer and Smith 2020; Lewicka, Siemion, and Kurcok 2015; Tyagi et al. 2021). In order to present a systematic discussion, the most common starch modification methods will be classified into chemical, physical, enzymatical, genetical, and their combinations (Figure 4) (Kaur et al. 2012).

#### ***4.1. Chemical modification of starch***

Starch chemical modification converts the functional groups into starch (Alcázar-Alay and Meireles 2015). Regarding chemical modification, different properties can be achieved as a function of the location of the hydroxyl group, bond type, and grain size (Lewicka, Siemion, and Kurcok 2015). In this sense, on an industrial scale, the most frequent chemical modifications comprise reactions of cationization, cross-linking, dextrinization, esterification, etherification, grafting, hydrolysis, oxidation, ozonation, among others. Of these, oxidation, esterification, and etherification are the main modification reactions used (P. Tomasik and Gładkowski 2001).

Oxidation, the most traditional modification method, introduces carbonyl and carboxyl groups to form hydrogen bonds with the hydroxyl groups of the starch, which makes the polymer matrix more integrated and consequently improving the mechanical properties of the oxidase starch film (Zavareze et al. 2012). The most common oxidizing agents are air or oxygen (in the presence of catalysts), inorganic peroxides (e.g.,  $H_2O_2$ ), organic peroxides (e.g.,  $NaClO$ ,  $NaIO_4$ ), nitrogen compounds (e.g.,  $HNO_3$ ,  $N_2O_4$ ), and organic oxidants (e.g.,  $CrO_3$ ) (Piotr Tomasik and Schilling 2004). Oxidized starches show physicochemical properties such as reduced viscosity, high clarity and low-temperature stability resulting in better applications in food (Singh, Kaur, and McCarthy 2007). Moreover, the chemical reaction time in oxidized starches influences the surface morphology and consequently their functionalization which results in a controlled biodegradation of the starch matrix (Sangseethong, Termvejsayanon, and Sriroth 2010; Lauer and Smith 2020).

The hydrophilicity of the starch deteriorates the mechanical properties and dimensional stability. Thus, esterification modification is the reaction of the three hydroxyl groups of starch converted into hydrophobic ester groups under the influence of organic and

inorganic acids and their derivatives (such as acid anhydrides, oxychlorides, chlorides) to improve the hydrophobicity, processability and flexibility and consequently the thermoplasticity (Lewicka, Siemion, and Kurcok 2015; Masina et al. 2017; Wang et al. 2020).

Etherification of starch is the substitution of hydroxyl groups in glucose by carboxyl methyl, hydroxypropyl and/or other modified groups forming an ether. However, starch hydrophobic modification by etherification (detailed mechanism) is still unexplored (Masina et al. 2017; Wang et al. 2020). In the last decades, major developments have been made in starch modification, particularly physical, enzymatic, and genetic modifications (starch source), as briefly described below.

#### ***4.2. Physical modification of starch***

The physical modification of starch is considered safer when compared to chemical modification. It includes cold plasma, deep freezing, heating, hydrothermal modification, mechanical modification, pH modification, high-pressure treatment, pulsed electric fields, radiation modification, ultrasound, dry heating treatment, among others (Yu et al. 2021). Native starches thermally treated in the presence of plasticizers reduce the melting temperature to below decomposition, making it possible to model them into a determined shape (Jiménez et al. 2012). Thus, heating processes can significantly impact the starch's physical properties (Maniglia, Castanha, Le-Bail, et al. 2021). Interest in the physical modifications of starch has grown in recent times, particularly in radiation as well as high and low-temperature treatments.  $\gamma$ -irradiation is a fast process that doesn't use high temperatures or produce chemical residues and is used to modify different starch sources, making it suitable to produce biodegradable packaging films (Bhat and Karim 2009; Zhu 2015; Teixeira et al. 2018; Kanatt 2020). Cold plasma irradiation, a novel technology for food packaging, has been used to produce a hydrophobic starch film with improved physicochemical properties using Sulfur Hexafluoride, 1-butene, Hexamethyldisilazane, Hexamethyldisiloxane, Argonium, and Helium (Hwang et al. 2005; Sheikhi et al. 2020).

#### ***4.3. Enzymatic modification of starch***

Although enzymatic methods are rarely used due to costs, complexity, and time-consumption, they present several advantages compared to chemical modification in that they are safer, attain higher yields, and are considered an environmentally friendly process (Yu et

al. 2021; Tibolla et al. 2019). Enzymatical modification, which uses enzymes to catalyze starch in enzymatic modification, including gelatinization, liquefaction, saccharification, debranching, transglycosylation, and isomerization reactions, decreases the solubility and improves the thermal, chemical, and mechanical properties of the films (Yu et al. 2021; Rosseto et al. 2021). The enzymatic reaction occurs in the amorphous regions, such as amylose and the branch points of amylopectin (A.O. Ashogbon 2021). Enzymes including transglutaminase, xylanase, transglucosidase, maltogenic  $\alpha$ -amylase,  $\beta$ -amylase and pullulanase are frequently used in starch modification to achieve improved rheological properties and increase its applicability in the food processing industry (Yu et al. 2021; Tibolla et al. 2019; Rosseto et al. 2021; Guo et al. 2018; Li et al. 2020).

#### ***4.4. Genetic modification of starch***

Genetic modification of starch through traditional plant breeding techniques or by using biotechnological methods involves enzymes related to starch biosynthesis. This process is of major interest since it changes the properties of the starch for specific purposes or new applications (Kathuria, Gautam, and Sharma 2019). Genetic modification generally focuses on the obtention of high amylose starch promoting more rigid and crystalline films (Kathuria, Gautam, and Sharma 2019; Luchese et al. 2018b; Cano et al. 2014; Colussi et al. 2017; Lauer and Smith 2020). Several genetic and molecular biological techniques have been developed to increase the amylose content. A mutation in the gene that encodes starch-branching enzyme IIb (amylose extender) produces high-amylose starch (Neelam, Vijay, and Lalit 2012). The use of single domain antibodies against starch-branching enzyme II was evaluated and produced higher amylose content starches (Jobling et al. 2003).

#### ***4.5. Sequential starch modification methods***

The use of a combination of modification methods to produce modified starch with specific functional properties in a short period of time and an increase in production and even to improve edible coatings/films to be used as food packaging (U. Shah et al. 2016) has grown in recent years (Kaur et al. 2012). Oyeyinka et al. (2021) detailed some impacts on the structure and physicochemical properties of starches, when treated with microwave heating alone and combined with other starch modification methods, such as carboxymethylation, retrogradation and conductive heating. The authors observed that microwave heating alone impacts the

amylopectin chain, disarranging the starch structure, while increasing amylose-amylose interactions, which makes the starch bonds stronger. Regarding the combination of microwaves with other methods, a general improvement in the functionality of the starch was observed, i.e. the reduction of the gelatinization temperature in corn starch-modified by microwave in combination with xanthan gum and the alteration of the crystalline pattern (from type B crystalline polymorph to type C) of potato starch modified by microwave followed by esterification (Oyeyinka et al. 2021). Similarly, Woggum, Sirivongpaisal, and Wittaya (2014) used two methods in sequence to modify rice starch (hydroxypropylated followed by crosslinking with sodium trimetaphosphate alone and mixed with sodium tripolyphosphate). The modified rice starch was then used to produce of biodegradable films. Among the main results, it was found that films with dual-modified rice starch increased the resistance (6.45–8.17 MPa of TS), the flexibility (96.98–126.11% of EB), the barrier property ( $6.96\text{--}7.63 \times 10^{-8} \text{ g m}^{-1} \text{ h}^{-1} \text{ Pa}^{-1}$  of WVP), and solubility of the films (6.29–7.86%), when compared to native rice starch-based film (6.18 MPa of TS, 68.7% of EB,  $1.24 \times 10^{-7} \text{ g m}^{-1} \text{ h}^{-1} \text{ Pa}^{-1}$  of WVP, and 4.45% solubility). It was also observed that the modified starch reduced the crystallinity and opacity of the films (Woggum, Sirivongpaisal, and Wittaya 2014).

Adeleke Omodunbi Ashogbon and Akintayo (2014) suggest that the main dual modifications involve a combination of chemical and physical or enzymatic and chemical methods, such as cross-linking/acetylation, acetylation/oxidation, or cross-linking/hydroxypropylation, in addition to ultra-high pressure-assisted/acetylation or high hydrostatic pressure-assisted/phosphorylation. A.O. Ashogbon (2021) states other types of dual modifications, for example, two chemicals (e.g. succinylation/cross-linking), two physical (e.g. extrusion/annealing), and two enzyme (e.g.  $\alpha$ -amylase/pullulanase) modifications. Some starch modification technologies, such as ozonation, are more prepared for implementation in industries, while others, such as pulsed electric field processing, still require further research (Maniglia, Castanha, Rojas, et al. 2021). It is important to emphasize that further studies should investigate the effect of combining technologies on the molecular structure and properties of starches in order to predict possible applications (Maniglia, Castanha, Le-Bail, et al. 2021).

## **5. Production of starch-based films**

Adequate conditions in the film production process are required to attain a cohesive matrix with a homogeneous structure and good mechanical and barrier properties. Production

methods act directly in the crystallization, melting, decomposition, granular expansion, and water diffusion phenomena. The standard processes used for starch films production are casting (Moraes et al. 2015; Thakur et al. 2019) and extrusion (Ceballos et al. 2020; Gao et al. 2021). Casting consists of preparing a liquid solution through heating, which the starch granule is gelatinized, followed by drying to form the final film. This method is commonly used to study starch gelatinization, plasticization, and the thermomechanical properties of the starch film, nevertheless it is not feasible on an industrial scale (Zhang, Rempel, and McLaren 2014). Extrusion uses pressure and temperature in a low moisture film matrix to form pellets, which are then converted into films by blowing, thermo-compression, or injection (Fakhouri et al. 2013; Suhag et al. 2020; Cheng et al. 2021). Figure 5 illustrates the main techniques for starch film production and Table 2 presents the main properties of starch-based films applicable for food packaging development by a different production technique.

Bench-casting involves casting the solution in a mold. It is the most commonly employed method in starch film development. This technique is widely used as it is simple, fast, and cheap equipment can be used. However, low productivity and up-scale difficulties are limitations (Fakhouri et al. 2013; Suhag et al. 2020; Cheng et al. 2021). Casting can obtain higher productivities by using the tape-casting technique. In tape-casting, the film solution is spread in large supports or on continuous carrier tapes, on which the resulting film is dried (Moraes et al. 2015; 2013; He et al. 2014). This is how the film properties promoted by bench-casting can be attained, but with increased productivity and thickness control. Casting has the advantage of promoting good interaction between particles, thus producing homogeneous films with few defects (Suhag et al. 2020). When using aqueous solutions, the films tend to present a higher moisture content compared to extruded film (Ochoa-Yepes et al. 2019). Water acts as a plasticizer as it reduces the hydrogen bonding between polymer chains, which increases the molecular volume and consequently the water vapor diffusion in the films (Ochoa-Yepes et al. 2019; Gao et al. 2021). This lowers the TS and YM and enhances EB (Ochoa-Yepes et al. 2019; Thakur et al. 2019). These attributes are linked to a cohesive matrix, indicated by the superficial homogeneity and absence of cracks or pores. Due to the high integrity and cohesiveness of casted starch films, their barrier and mechanical properties are relatively good when compared with porous or cracked films often formed by extrusion (Fakhouri et al. 2013). Figure 6 presents average mechanical properties (TS, EB, and YM) of starch-based films from different sources (native or modified starch) (Tang et al. 2022; Gutiérrez and Valencia 2021; Narváez-Gómez et

al. 2021; Żołek-Tryznowska et al. 2021; Costa et al. 2021; Silvia et al. 2021; Sifuentes-Nieves et al. 2021; Ceballos et al. 2020; La Fuente et al. 2020; Estevez-Areco et al. 2019; Ochoa-Yepes et al. 2019; Dai et al. 2019; Ibrahim et al. 2019; Luchese et al. 2018a; Tirado-Gallegos et al. 2018; González-Soto et al. 2018; Colussi et al. 2017; Colivet & Carvalho 2017; Moraes et al. 2015).

On a commercial scale, extrusion is the most used method due to its relative low-cost production and high productivity compared to the casting method (Suhag et al. 2020; Cheng et al. 2021). This process applies mechanical and thermal energy to the material, which can reach high temperatures at low residence times. The screw speed, pressure at the die, energy input, and thermal profile inside the extruder are some of the possible parameters. Extrusion has the advantage of being a low moisture, and energy consumption process that can be carried out quickly. Extrusion also offers good control of the resulting film's properties and versatility of forms (Suhag et al. 2020). Additionally, aiming to enhance the properties of the film, it can be associated with reactions that aim to modify the polymer, such as grafting, cross-linking, polymerization, and poly-condensation (Cheng et al. 2021; Gutiérrez and Valencia 2021). Extrusion processes are, however, not suitable for thermo-sensitive and high moisture material blends. The high cost of equipment and maintenance could also be a limitation of this technique (Suhag et al. 2020).

As it is a dry method, well-structured extruded starch films tend to have a lower moisture content than films produced by casting (Ochoa-Yepes et al. 2019). This leads to a lower plasticizer effect by the water. Thus, hydrogen bonding between molecules from the matrix are stronger, leading to lower solubility and WVP. For the same reason, extruded films may also present thermoplastic behavior, but with a higher YM and TS, and a lower EB. Extruded starch films are generally more opaque than casted films, which could be caused by higher crystallinity or structural changes induced by the process (Fakhouri et al. 2013). As extrusion uses high pressures, temperatures, and shear rates, it may reduce the crystallinity of starch, promoting the formation of heterogeneous, brittle, and rough films with poor mechanical properties (Fakhouri et al. 2013; Cheng et al. 2021). Cracks and pores are a consequence of the stiffness, which can harm the barrier and mechanical properties (Fakhouri et al. 2013).

## **6. Perspectives and conclusion**

Food packaging is essential for maintaining food quality and safety, also must contribute for food waste reduction. Consumer demand for safe, high-quality food products, greater convenience, and ease handling is growing. Furthermore, greater environmental awareness, concern, and pressures of the community has encouraged industries and governments to meet sustainability goals that go beyond reducing petrochemical plastics to replacing them with new materials. Large food industry companies are increasingly expected to take initiatives towards a more sustainable economy, with the adoption of circular economy principles and modernizing their packaging, especially after the impacts caused by the COVID-19 pandemic.

Furthermore, biodegradable packaging for food is encouraged and valued, boosting the market for biopolymers, especially those from biomass. The use of biopolymers extracted from agro-industrial waste, can contribute to reducing food loss and to producing more sustainable materials, adding value to waste that would otherwise be discarded. Another advantage of using food waste in biodegradable films is associated with its potential to provide bioactive compounds for packaging, making them active and/or intelligent. These actions represent a future trend given that global goals involve the adoption of systems based on the reuse of raw materials and other strategies to achieve the global environmental goals established in the 2030 Agenda.

Therefore, the most promising research perspective on biopolymer-based films, especially starch, should investigate the scale production drawbacks, reduce costs, improvement on mechanical and barrier properties, among other. In addition, it is important to emphasize that biopolymer-based films must be evaluated in terms of safety and toxicity, to contribute to legal regulation.

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### **Author contributions**

Matheus, JRV, Fai, AEC, and Andrade, CJ designed the study; Matheus, JRV, Dalsasso, RR, Rebelatto, EA, Andrade, KS, and Andrade, LM, wrote the paper; Fai, AEC, Andrade, CJ, and Monteiro, AR provided critical revision of the article; Fai, AEC supervised the project.

### **Conflict of interest**

There are no conflicts of interest.

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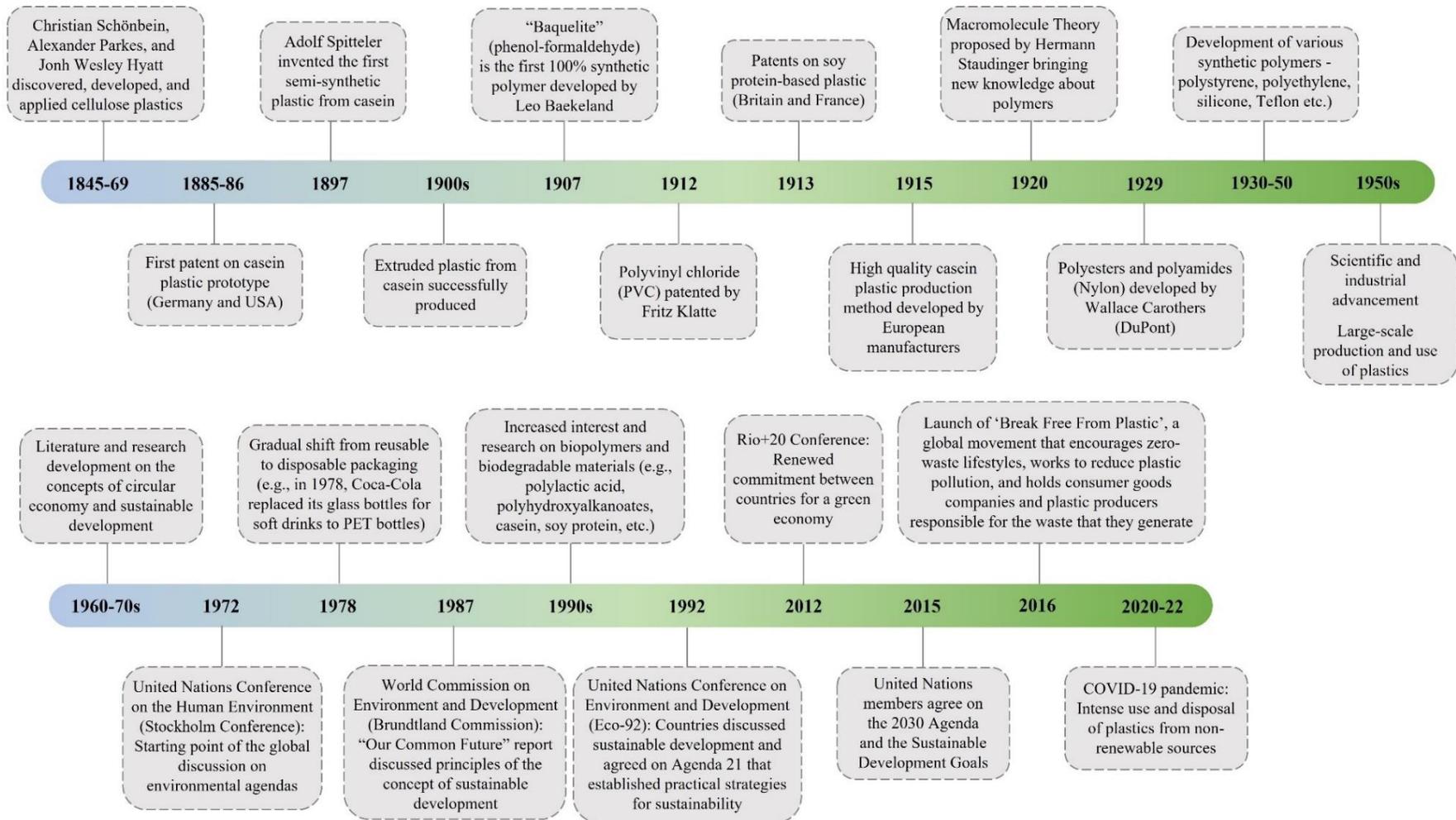
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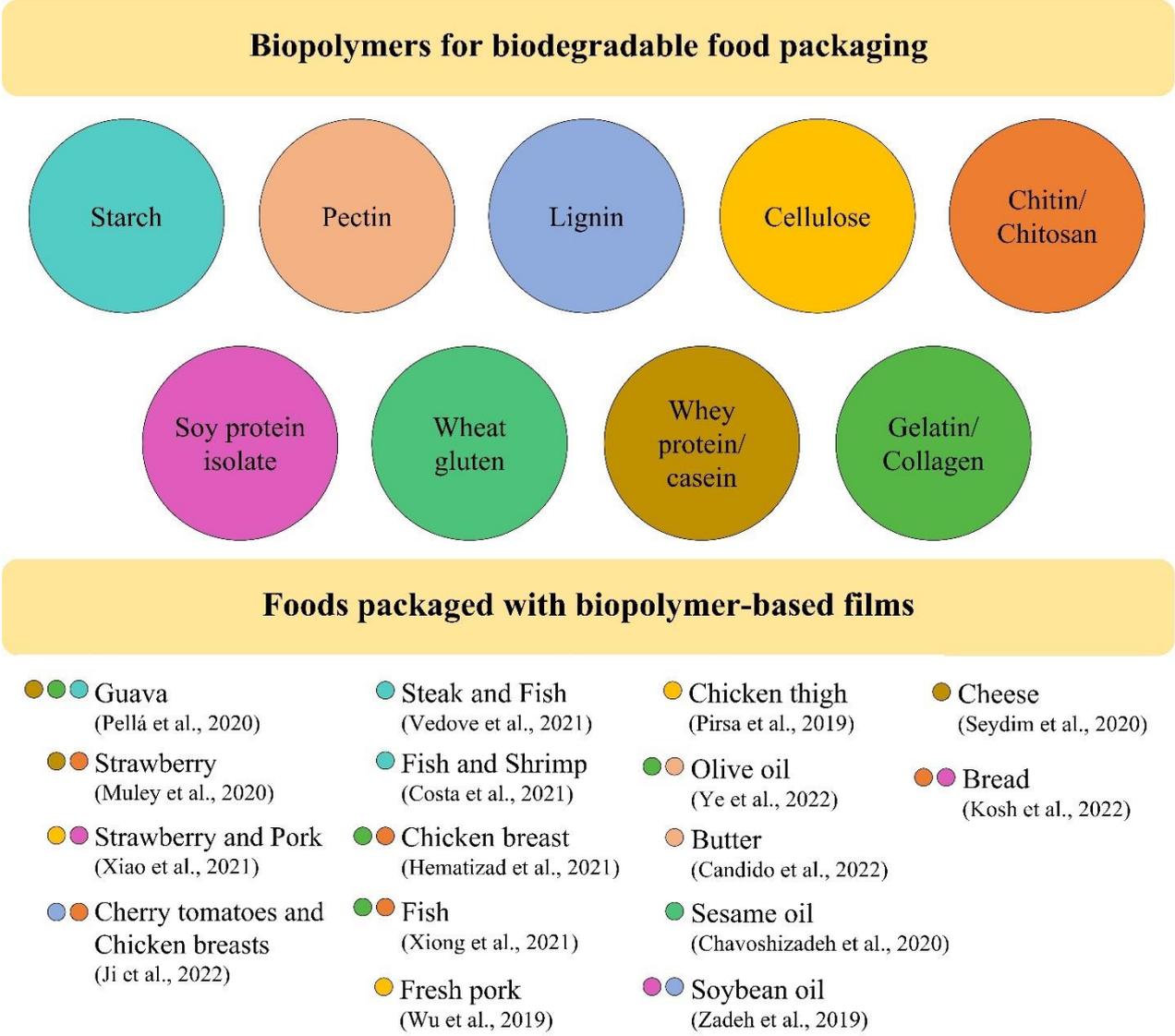
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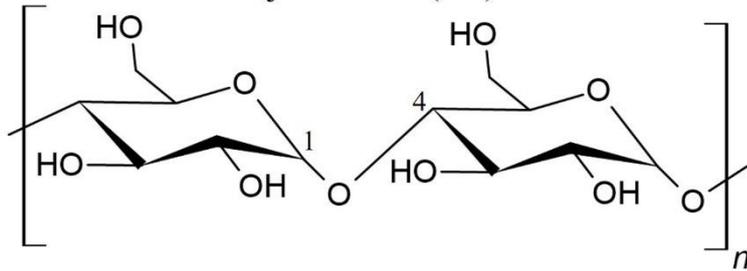
**Figure 1.** Timeline highlighting the main events and discoveries on plastics, bioplastics and world conferences on the environment and sustainability (UN 2015; Arkin et al. 2019; Osswald and García-Rodríguez 2011; Ignacio 2020; Velenturf and Purnell 2021).



**Figure 2.** Primary biopolymers from biomass and agro-industrial residues used for the development of biodegradable food packaging.

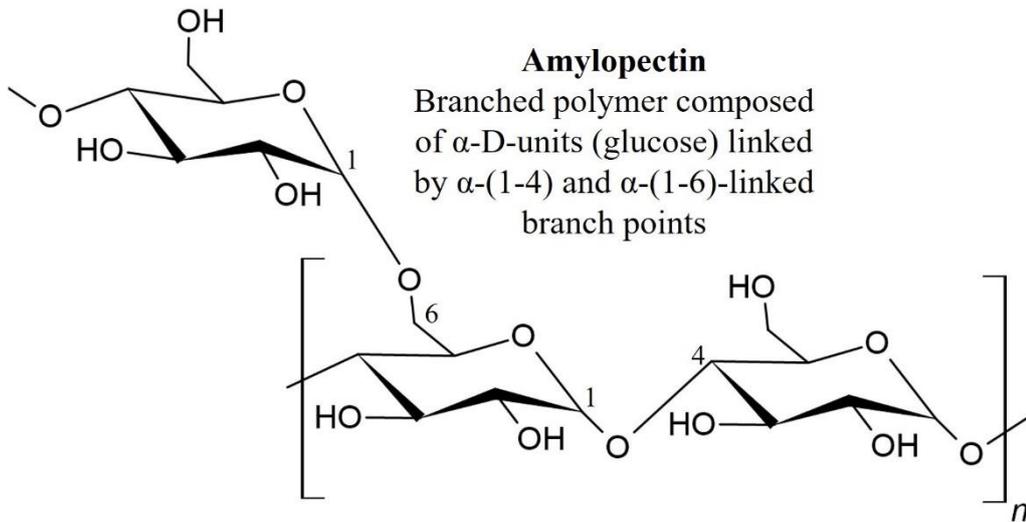
### Amylose

Linear polymer composed  
of  $\alpha$ -D-units (glucose)  
joined in  $\alpha$ -(1-4)



### Amylopectin

Branched polymer composed  
of  $\alpha$ -D-units (glucose) linked  
by  $\alpha$ -(1-4) and  $\alpha$ -(1-6)-linked  
branch points



**Figure 3.** Chemical structure of starch: amylose and amylopectin.

### Chemical modification

Introduces functional groups into the starch molecule by derivatization reactions (e.g., esterification, etherification, oxidation, ozonation, cross-linking, cationization, dextrinization, grafting, hydrolysis, etc.)

*Example:* Ramírez-Centeno et al. (2021) studied the effect of chemical modification of banana starch through starch reactions with virgin PET and PET bottle waste and its influence on film properties.

### Physical modification

Can be classified into thermal (e.g., deep freezing, heating, hydrothermal, dry heating treatment, etc.) and non-thermal modification (cold plasma, mechanical, pH, pressure, pulsed electric fields, radiation, ultrasound, etc.)

*Example:* Bangar et al. (2021) evaluated the influence of different physical treatments (heat-moisture, microwave, and sonication treatment) on pearl millet starch and studied these modified starches for film development.

### Enzymatical modification

Modification of starch molecule using enzymes, such as transglutaminase, xylanase, transglucosidase, maltogenic  $\alpha$ -amylase,  $\beta$ -amylase, and pullulanase

*Example:* Miao et al. (2021) developed an active food packaging film based on corn starch modified by enzymatic hydrolysis and incorporating tea polyphenols. This modification generated a porous starch that, when applied in the film formulation, favored its properties of slow release of bioactive compounds.

### Genetic modification

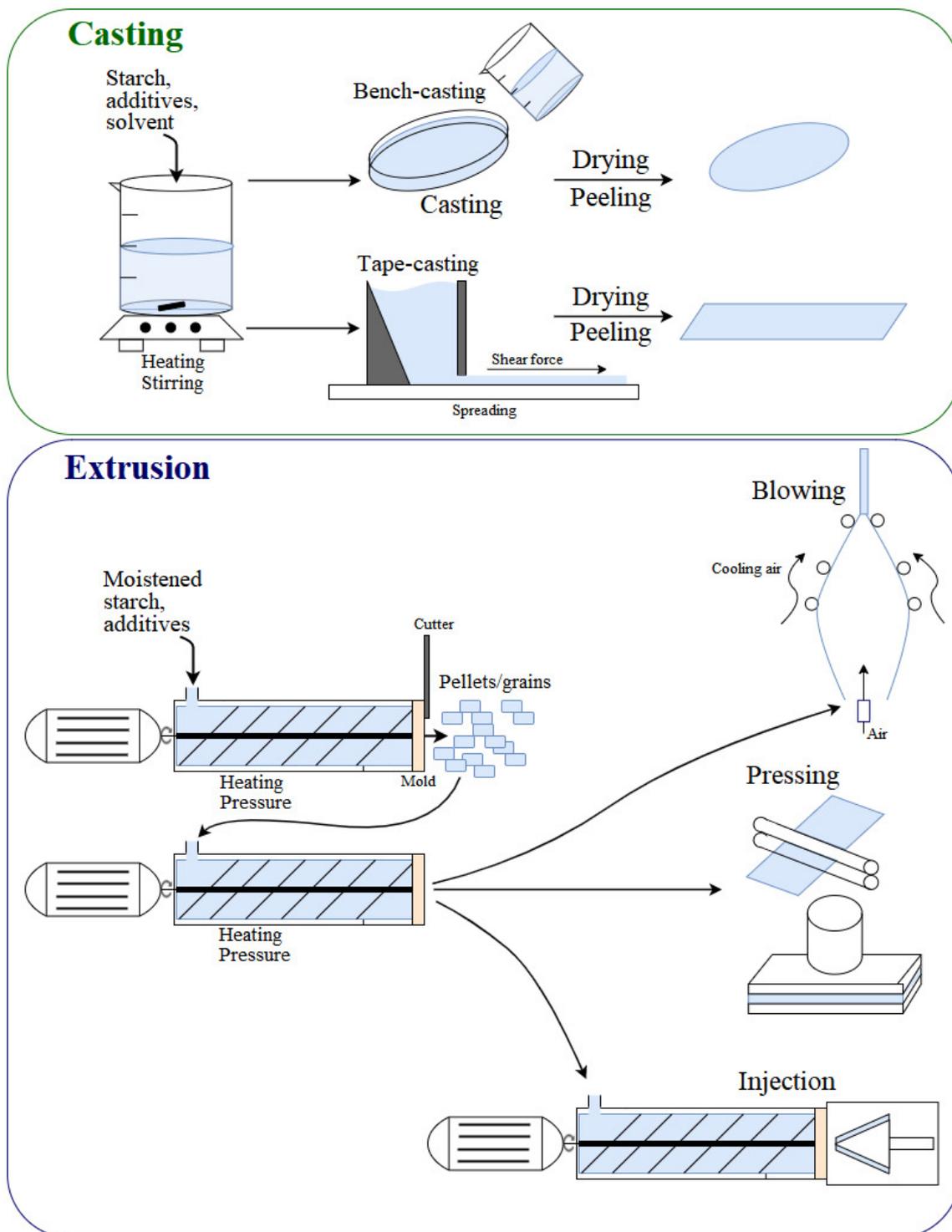
Can be done by traditional plant breeding techniques or by using biotechnological methods involving enzymes

### Sequential modification

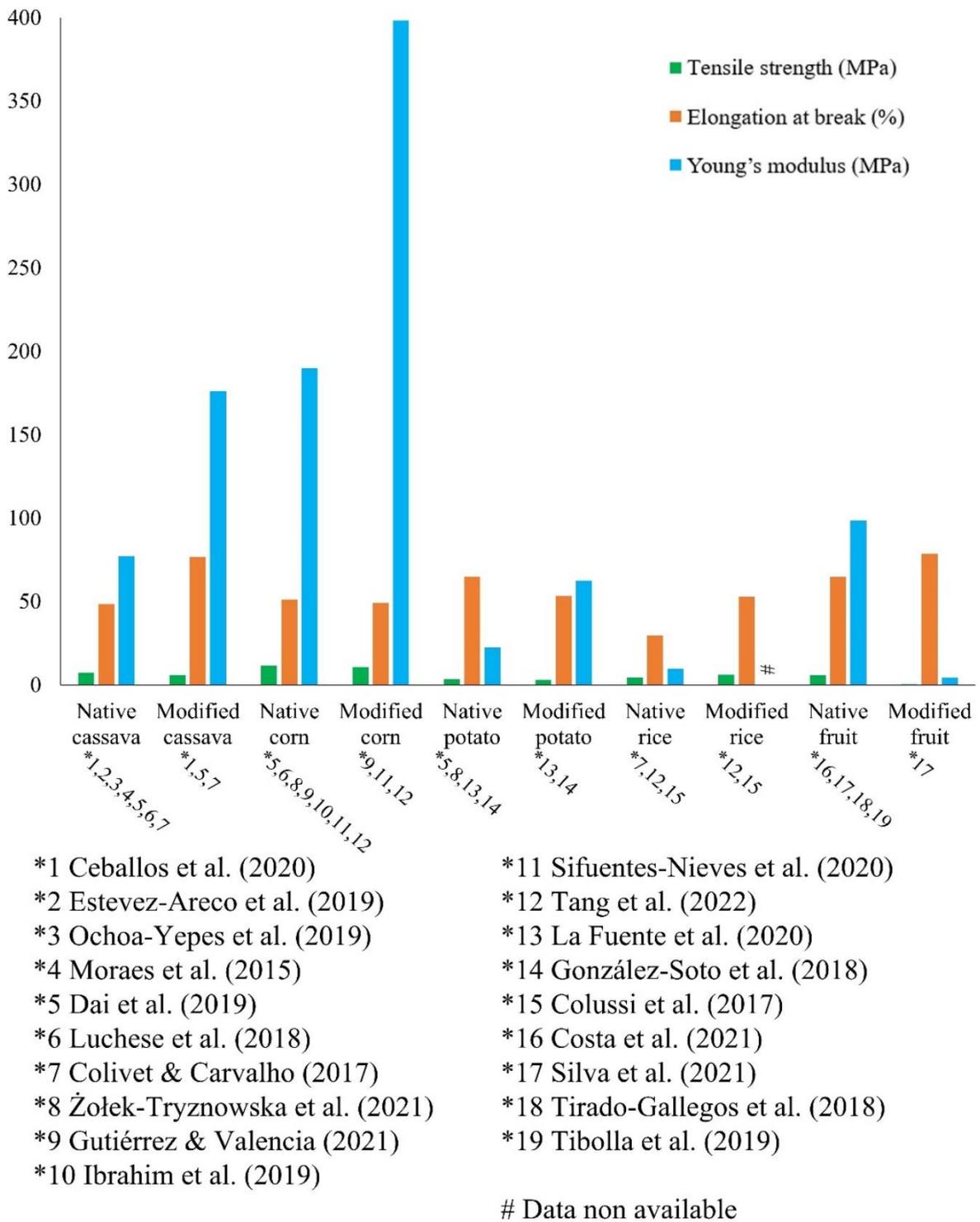
Various combinations of methods for modifying starch are possible, such as physical/chemical (e.g., ultra-high pressure-assisted/acetylation, high hydrostatic pressure-assisted/phosphorylation); enzymatic/chemical (e.g.,  $\alpha$ -amylase/acetylation); two physicals (e.g., extrusion/annealing); two chemicals (e.g., succinylation/cross-linking; cross linking/acetylation, acetylation/oxidation, or cross linking/hydroxypropylation); and two enzymatical methods (e.g.,  $\alpha$ -amylase/pullulanase)

*Example:* Narváez-Gómez et al. (2021) developed films of modified yam starches (oxidized, cross-linked, and dual-modified) evaluating their impact on mechanical, optical, structural and barrier properties.

**Figure 4.** Chemical, physical, enzymatical, genetical methods, and combinations of starch modification.



**Figure 5.** Main starch film production techniques.



**Figure 6.** Mechanical properties (average TS, EB, YM) of starch-based films from different sources (native or modified).

**Table 1.** Main properties and outcomes of recent studies on the development of biopolymer-based packaging materials.

Main biopolymer (source)	Film-formulation additives	Mechanical properties	WS (%) WVP (g m <sup>-1</sup> h <sup>-1</sup> Pa <sup>-1</sup> )	Bioactive properties	Outcomes	Ref.
Soy protein isolate (soybean)	Silver nanoparticles Chitin nanowhisker	TS 4.7–16.2 EB 7.1–83.0% Th 157.0–253.0 μm	WVP 1.9–9.9 x 10 <sup>-6</sup>	Antimicr. (halo size <sup>*1</sup> ): <i>E. coli</i> 13 mm, <i>B. cereus</i> 8 mm. Antiox.: 2.5–27.5 % DPPH scavenging ability.	Additives contributed to increase mechanical strength, barrier properties, in addition to increasing the bioactivity of the films compared to the soy protein isolate film. However, these improvements in the properties of the films impacted on the reduction of the biodegradability rate, with the control film reaching 95% degradation after 24 days of analysis and the films with the additives ranging from 80-90%. Bread pieces packed with a soy protein isolate film with nanoparticles showed lower fungal growth and remained fresh until the 15th day of storage, unlike breads packed with film without additives and with polyethylene, which already showed signs of fungal contamination on the 4th day.	Kosh et al. 2022

Soy protein isolate (soybean)	Cellulose nanocrystals (CNC)	TS 4.5–5.5 MPa EB 75.0–140.0% Th 74.0–81.0 $\mu\text{m}$	WS 20.0–32.0 WVP 7.2– $12.0 \times 10^{-7}$	-	Higher TS and lower WVP values were found for films with 0.5 and 0.75% CNC in the formulation. However, high concentrations of CNC probably agglomerated and negatively impacted mechanical strength and barrier property. The EB values decreased as the CNC concentration increased. The application of the films as fresh pork packaging resulted in visible microbial colonies in the negative control (uncovered) and the sample covered with film without CNC after 6 days of refrigeration, suggesting that CNC incorporated in the films was able to inhibit microbial growth. Films containing CNC had better effects in delaying weight loss of strawberries than uncovered samples.
Soy protein concentrate (soybean)	Free and microencapsulated oregano essential oil	YM 41.4–375.4 (178.7) MPa TS 2.5–19.9 (10.0) MPa EB 26.1–259.5 (177.7)% Th 210.0–327.0 (286.3) $\mu\text{m}$	WS 39.3–61.5 (47.5) WVP 3.1–7.5 (5.5) $\times 10^{-7}$	Antimicr. (halo size <sup>*1</sup> ): <i>E. coli</i> 6.3–10.7 mm, <i>S. aureus</i> 14.2–31.7 mm. TPC: 1.3–56 mg gallic acid/g film. Antiox.: 2967–11000, 10293–760000, and 3842–7250 $\mu\text{M}$ trolox equivalent/ g film by DPPH, ABTS, and FRAP method, respectively.	The addition of microencapsulated essential oil made the films stronger (19.9 MPa of TS and 375.4 MPa of YM) and with better barrier properties ( $3.1 \times 10^{-7} \text{ g m}^{-1} \text{ h}^{-1} \text{ Pa}^{-1}$ of WVP) compared to the free essential oil film and the control film. Furthermore, the film with microencapsulated essential oil was shown to be superior in terms of antimicrobial properties against <i>E. coli</i> and <i>S. aureus</i> . Free essential oil was only more favorable than microencapsulated essential oil in the antioxidant aspect of the soy protein-based film.

Soy protein isolate (soybean)	Licorice residue extract	TS 7.7–10.83 MPa EB 100.5–170.0%	WVP 6.3– $7.3 \times 10^{-7}$	TPC: 1.4–5.6 mg gallic acid/g film. Antiox.: 3.0–57.0 % DPPH scavenging ability, 12.2–82.9 % ABTS scavenging ability.	The incorporation of licorice residue extract decreased the WVP and increased the TS of the films. The films released more total phenolic content and antioxidant into alcoholic and fatty food simulants compared with control films (without extract) and possessed stronger DPPH and ABTS scavenging activities.	Han et al. 2018
Soy protein isolate (soybean)	Soybean straw nanofibrils or nanocrystals	YM 459.0–575.0 (523.7) MPa TS 6.1–9.0 (7.8) MPa EB 4.2–18.0 (10.1)% Th 62.0–82.0 (74.7) $\mu\text{m}$	WS 20.0–33.0 (26.3) WVP 2.5–5.0 (4.0) $\times 10^{-6}$	-	The addition of nanoparticles in the soy protein-based film increased the thickness (80–82 $\mu\text{m}$ ), strength (8.4–9.0 MPa of TS), and moisture content (17%), reducing the flexibility (4.2–8.0% of EB). The film with soybean straw nanofibrils stood out with the best barrier property ( $2.5 \times 10^{-6} \text{ g m}^{-1} \text{ h}^{-1} \text{ Pa}^{-1}$ of WVP) and a 48% increase in TS compared to the control film.	Martelli-Tosi et al. 2018
Collagen (fish)	Willow bark dry extract	YM 627.0–1420.0 MPa TS 41.7–60.7 MPa	-	-	The addition of extract modified the roughness of collagen films and improve their mechanical properties, increasing TS and YM.	Adamiak et al. 2021
Collagen (bovine)	Micro and nano collagen fibers	TS 14.0–92.4 MPa EB 2.3–4.4% Th 31.0–122.0 (44.0) $\mu\text{m}$	WS 15.5–27.0 (16.9)	-	The films prepared with nanofibers had a more uniform and denser structure, mainly attributed to the physical entanglement and non-covalent bonds, which favored improvements in the mechanical and the water barrier properties of the films.	Ma et al. 2020
Collagen (bovine)	Thyme essential oil	TS 18.3–38.5 (27.3) MPa EB 23.5–48.6 (36.3)% Th 12.7–23.0 (17.7) $\mu\text{m}$	WS 74.3–87.6 (79.2)	Antiox.: 3.9–53.2 (30.9) % DPPH scavenging activity.	Essential oil added collagen-based films presented an increase of 1.8x in thickness (0.23 mm), 2.1x in EB (48.56%), 13.6x DPPH radical scavenging capacity (53.15%), and a low barrier performance; and a decrease of 0.5x in TS (18.25 MPa) and 0.8x in film solubility (74.3%), when compared to the control film (only collagen).	Ocak 2020

Collagen (bovine)	Laponite	YM 62.7–81.9 (69.8) MPa TS 9.2–11 (10.1) MPa EB 42.3–53.8 (49.2)% Th $\pm$ 70.3 $\mu$ m	WVP $\pm$ 4 x 10 <sup>-7</sup>	-	The incorporation of different concentrations of nanoparticles of laponite in collagen-based film did not affect the TS (9.2–11 MPa), EB (42.3–53.8%) or YM (62.7–81.9 MPa). Neither the WVP ( $\pm$ 4 x 10 <sup>-7</sup> g m <sup>-1</sup> h <sup>-1</sup> Pa <sup>-1</sup> ) nor the water contact angle ( $\pm$ 89 °) were affected by the incorporation of laponite, suggesting that films are able to retain hydrophobic surfaces.	Valencia et al. 2019
Gelatin (bovine)	Zinc oxide nanoparticle Mentha piperita essential oil	YM 1626.7– 3037.0 (2389.9) MPa TS 48.2–61.9 (56.0) MPa EB 7.7–11.5 (9.2)% Th 107.0–144.0 (128.0) $\mu$ m	WS 32.9– 42.5 (36.7)	Antimicr. (halo size*1): <i>E. coli</i> 9.0– 98.0 mm.	The combination of additives incorporated in the gelatin film reduced WS, WVP and EB of the films, while the thickness, TS and YM increased. In addition, the developed films showed good antibacterial activity against <i>Escherichia coli</i> O157:H, associated with the synergistic effect of the additives in gelatin films.	Javidi et al. 2021
Gelatin (chrome leather scrap)	$\beta$ -cyclodextrin	TS 65.7–122.3 (86.2) MPa EB 6.5–15.7 (9.9)%	-	-	The mixtures of $\beta$ -cyclodextrin and gelatin (ratio 1:2) were compatible and formed a film with good mechanical properties (122.34 MPa of TS; 15.74% of EB), and thermal stability up until 319 °C, as well as high biodegradability (about 80% in 100 days).	Dang, Shan, and Chen 2018

Gelatin (bovine)	Durian leaf extract	-	WVP 4.1–7.5 (5.8) x 10 <sup>-4</sup>	Antiox.: 0.3–0.7 and 0.7–14.1 mg/mL TE/100 mg film by DPPH and FRAP method, respectively.	The extract did not improve the water barrier properties of the gelatin-based film. The film with 0.5% of extract increased by 18x the DPPH radical scavenging capacity and was 3x more able to carry out the retardation of palm oil oxidation than the negative control sample. There was no difference between the values of retardation of oil oxidation of film added with 0.5% extract and commercial plastic, which is considered promising given the biodegradable advantages of gelatin-based film. The gelatin-based film did not affect the changes in weight and texture of the durian fruit pulps during storage and represented the most efficient commercial plastic.	Kann et al. 2018
Gluten (wheat)	Chlorophyll	TS 45.0–95.0 MPa EB 197.0–275.0%	-	Antiox.: 60.0–93.0%.	Film enhanced the shelf life of sesame oil, decreasing peroxide value in oil containing film compared to filmless oil. Also, the chlorophyll pigment improved the properties of gluten film and had high potential for improving food shelf life and can be used as a smart sensor for detection of the expiration date of sesame oil.	Chavoshizadeh et al. 2020
Gluten (wheat)	Zinc oxide nanoparticle	TS 100.0–230.0 MPa EB 260.0–315.0%	-	-	The increase in the content of gluten in the formulation resulted in greater TS, EB, and water absorption, but lower solubility of the films. Furthermore, gluten did not present an association with the antimicrobial and antioxidant activities of the films. On the other hand, the increase in zinc oxide in the formulation generated gluten-based films with greater antimicrobial and antioxidant properties, without affecting the values of the TS, yet increasing the EB values.	Rezaei, Pirsā, and Chavoshizadeh 2020

Gluten (wheat)	Xanthan gum Glyoxal	YM 5.3–20.0 MPa TS 0.5–1.5 MPa	-	-	Extrusion resulted in more resistant films compared to production by compression, given the greater gluten-plasticizer compatibility. The use of a more alkaline pH positively impacted the water uptake capacity and TS of gluten-based films. Xanthan gum and glyoxal produced films with less deformation and swelling capacity.	Jiménez-Rosado et al. 2019
Chitosan (commercial)	Lemongrass essential oil	TS 7.9–15.9 (11.2) MPa EB 32.5–65.3 (47.6)% Th 40.3–52.8 (47.2) $\mu\text{m}$	WS 5.2–21.8 (9.0) WVP 7.7–9.1 (8.3) x $10^{-5}$	Antimicr. (halo size <sup>*1</sup> ): <i>B. cereus</i> 5.7–8.3 mm, <i>E. coli</i> 7.3–11.3 mm, <i>L. monocytogenes</i> 6.3–11.3 mm, <i>S. typhi</i> 8.0–11.7 mm.	The addition of 9% essential oil in chitosan-based films increased thickness by 31% and EB by 101%; it also reduced water solubility by 76% and TS by 50% compared to the control film. The barrier property was not affected by the incorporation of essential oil. In addition, higher concentrations of essential oil added to the formulation made the film more opaque and increased antimicrobial properties, especially against <i>S. typhi</i> , followed by <i>L. monocytogene</i> , <i>E. coli</i> , and <i>B. cereus</i> .	Han Lyn and Nur Hanani 2020
Chitosan (commercial)	Nano-organoclay Zinc oxide nanoparticles	YM 1750.0–2410.0 (2095.0) MPa TS 18.3–38.9 (29.9) MPa EB 0.8–3.5 (1.7)%	-	Antimicr. (halo size <sup>*1</sup> ): <i>E. coli</i> 0.0–30.0 mm, <i>S. aureus</i> 0.0–20.0 mm. Antimicr. (% reduction by death curve test in 24 h): <i>E. coli</i> 95.4–100.0 %, <i>S. aureus</i> 99.5–100.0%.	The higher concentration of both nanoparticles in the chitosan-based film generated a synergistic effect in increasing strength. The thermal degradation temperature of the chitosan film reduced from 289 to 269 °C with the addition of zinc oxide nanoparticles. Regarding the antimicrobial property, a synergistic effect of chitosan and zinc oxide nanoparticles in the film against <i>E. coli</i> and <i>S. aureus</i> was observed.	Rodrigues et al. 2020
Chitosan (commercial)	Xanthan gum	TS 8.8–17.0 (12.9) MPa EB 23.5–53.1 (30.3)% Th 97.0–103.0 (100.7) $\mu\text{m}$	WS 21.6–22.6 (22.1) WVP 4.7–5.1 (4.9) x $10^{-7}$	-	The higher xanthan gum content in the chitosan film formulation had an impact on the increase in TS and in the reduction of EB, but it did not affect the WVP, WS, and moisture of the films.	De Moraes Lima et al. 2017

Cellulose (commercial)	Lignin	TS 44.5–92.0 (72.1) MPa	-	Antimicr. (halo size*1): <i>E. coli</i> 7.8–13.2 mm, <i>S. aureus</i> 7.8–10.7 mm.	The cellulose crosslinked film with lignin showed higher mechanical strength and barrier properties than the film without lignin. The addition of lignin also contributed positively to the antimicrobial activity and UV blocking capacity.	Lu et al. 2022
Lignocellulosic compounds (cassava bagasse)	Lignocellulosic nanofiber Nanoclay	TS 4.6–6.6 (5.4) MPa EB 43.8–54.9 (47.8)% Th 110.0–130.0 (120.0) μm	WS 6.4–31.3 (21.0) WVP 1.3–2.0 (1.7) x 10 <sup>-9</sup>	-	The composition of the lignocellulosic compounds was 27% of cellulose, 30% of hemicellulose, 2.7% of lignin. The lowest concentration of nanofiber in the formulation reduced the WVP and the solubility of the starch film. By increasing the concentration of nanofiber, greater thermal stability in the film was observed. Both nanoparticles were able to reinforce the starch film, reducing its flexibility. Furthermore, films with nanoparticles had lower opacity than the control film.	Travalini et al. 2019
Lignocellulosic compounds (corn husk residues)	Corn starch Fructose	YM 50.0–639.6 MPa TS 7.0–12.8 MPa Th 195.0–265.0 (235) μm	WS 20.5–23.3 (21.9)	-	The composition of the lignocellulosic compounds was 45.7% of cellulose, 35.8% of hemicellulose, 4.03% of lignin. The fibers reinforced the starch film, particularly in the formulation with the highest concentration tested, resulting in the highest values of TS and YM. The reinforced films, in comparison with the control film, showed greater thermal stability, as evidenced by the increase in the initial decomposition temperature (from 277 to 303 °C). Moreover, the higher concentration of fibers in the formulation increased the biodegradation rate of the film in the soil.	Ibrahim et al. 2019

Pectin (citrus)	Pracaxi oil nanoemulsions	-	WVP 0.8–3.2 x 10 <sup>-9</sup>	TPC: 56.7–83.2 (71.0) mg gallic acid/ 100g film. Antiox.: 23.3–50.3 (33.6) % DPPH scavenging activity.	Butter samples packed in films with or without nanoemulsions showed no difference in malondialdehyde values for 30 days, while PVC samples showed higher values of malondialdehyde. With 60 days of storage, films with nanoemulsions were more efficient to protect the butter samples from oxidation.	Candido et al. 2022
Pectin (lime peel)	Glycerol or coconut water Lime peel extract	YM 91.1–974.8 (578.4) MPa TS 3.6–20.6 (11.7) MPa EB 1.8–4.6 (3.2)% Th 48.3–49.8 (48.8) μm	WS 3.4–67.1 (44.3)  WVP 6.7–9.2 (7.4) x 10 <sup>-7</sup>	TPC: 0.0-81.0 (27.4) mg gallic acid/g film. Antiox.: 2.3-598.0 (155.3) and 5.5-52.0 (13.6) μM trolox/ g film by ABTS and DPPH assays, respectively.	Pectin-based films plasticized with coconut water were more flexible (2.31–4.78% of EB) and less soluble (27.26–36.61%) in water than those plasticized with glycerol. The addition of lime peel extract increases the solubility in the pectin films with both plasticizers (62.43–67.14%). Regarding the barrier property, it was observed that both plasticizers increased the WVP (6.7–9.2 x 10 <sup>-7</sup> g m <sup>-1</sup> h <sup>-1</sup> Pa <sup>-1</sup> ), but when coconut water and lime peel extract were added together in the filmogenic formulation it caused a reduction in the WVP (5.9–7.63 x 10 <sup>-7</sup> g m <sup>-1</sup> h <sup>-1</sup> Pa <sup>-1</sup> ). In addition, the lime peel extract provided greater antioxidant activity than the pectin film.	Rodsamran and Sothornvit 2019
Pectin (commercial)	Choline chloride	TS 3.0–18.4 (11.8) MPa EB 0.5–1.3 (0.8)% Th 240–400 (345) μm	WVP 5.2–9.8 (7.3) x 10 <sup>-6</sup>	-	The pectin film added with glycerol presented higher TS values (18.41 MPa) than the films containing plasticizers NADES (14.31 MPa) or ChCl (10.53 MPa), especially at the lower thermocompression time (20 min). Glycerol also favored the barrier property in the pectin film (5.2–5.6 x 10 <sup>-6</sup> g m <sup>-1</sup> h <sup>-1</sup> Pa <sup>-1</sup> of WVP) compared to the other plasticizers tested (6.6–9.8 x 10 <sup>-6</sup> g m <sup>-1</sup> h <sup>-1</sup> Pa <sup>-1</sup> of WVP).	Gouveia et al. 2019

Starch (cassava)	Cinnamon essential oil	TS 0.8–2.1 (1.4) MPa EB 3.5–42.2 (19.9)% Th 73.0–137.2 (105.7) $\mu\text{m}$	WS 18.7–52.8 (32.3) WVP 3.7–6.8 (5.4) $\times 10^{-7}$	-	The incorporation of essential oil in the starch film increased EB, water resistance, WVP and reduced TS.	Zhou et al. 2021
Starch (cassava)	Pectin Lemongrass essential oil	YM 975-1439 (1258) MPa TS 16.0–31.0 (23.4) MPa EB 2.4–4.7 (3.1)% Th 50.0–200.0 (108.0) $\mu\text{m}$	WVP 2.3–3.8 $\times 10^{-6}$	-	The incorporation of essential oil and pectin in starch film had a positive effect on the colorimetric parameters as well as the moisture barrier and thermal and mechanical properties, without affecting the rate of biodegradation in the soil ( $\pm 99.5\%$ of weight loss in 31 days). In general, the highest mechanical and barrier property values were found in the formulation containing 2% of pectin and 0.5% of essential oil.	Mendes et al. 2020
Starch (cassava)	Anthocyanin	YM 2.2–10.5 (4.4) MPa TS 0.3–1.8 (0.7) MPa EB 32.4–50.0 (38.8)% Th 600.0–1700.0 (1300.0) $\mu\text{m}$	WS 40.7–45.6 (42.4)	Films showed behavior as a smart packing indicator of pH change in meat stored at 6 °C.	The addition of anthocyanin to starch film reduced mechanical strength and stiffness by $\pm 4.5x$ and flexibility by $\pm 1.4x$ . However, starch film with anthocyanin, mainly in the highest concentration (20%), can be used as intelligent packaging for meat stored under refrigeration, with a noticeable change in color associated with pH ( $\Delta E^*$ ranging from 34.7-91.8, which indicates a most visually notable color change).	Vedove, Maniglia, and Tadini 2021
Polyvinyl chloride (PVC) based film (commercial)	-	YM 81.0 MPa TS 17.1 MPa EB 45.0% Th 8.0 $\mu\text{m}$	WVP 3.0 $\times 10^{-8}$	-	PVC, LDPE, and CS showed lower thickness, higher barrier properties compared to films developed based on starch (corn, cassava or mixture) and mixture of starch with chitosan (only film based on pure chitosan presented results statistically similar to commercial films). Regarding the mechanical properties, the PVC film presented a higher value of TS and a lower value of EB than most films and the CS film commercial presented a higher value of YM compared to the other analyzed films.	Luchese et al. 2018a
Low density polyethylene (LDPE) based film (commercial)	-	YM 71.0 MPa TS 3.9 MPa EB 99.0% Th 8.0 $\mu\text{m}$	WVP 3.0 $\times 10^{-9}$	-		

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Corn starch (CS) film (commercial)	-	YM 205.0 MPa TS 10.1 MPa EB 98.0% Th 1.1 µm	WVP 4.0 x 10 <sup>-8</sup>	-
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Values expressed as “minimum-maximum (average)”, when possible. Legend: “-” (information not available), Th (thickness), YM (Young’s modulus), TS (tensile strength), EB (elongation at break), WS (water solubility), WVP (water vapor permeability), Antimicr. (antimicrobial activity), Antiox. (antioxidant activity), TPC (total phenolic content), \*1 (inhibition halo size by agar diffusion test).

**Table 2.** Properties of starch-based films applicable for food packaging.

Starch source	Starch modification	Other components	Production technique	Mechanical properties	Barrier properties	Bioactive properties	Ref.
Cassava (18% amylose, 82% amylopectin)	Native	Yerba mate extract and water	Extrusion and compression	YM 42.0–63.0 MPa TS 2.8–3.0 MPa EB 60.0–74.0% TT 1.3–1.6 MJ m <sup>-3</sup> Th 300.0 μm	WVP 1.7–2.9 x 10 <sup>-6</sup> g m <sup>-1</sup> h <sup>-1</sup> Pa <sup>-1</sup>	AA 8.8–44.4 μM TE g <sup>-1</sup>	Ceballos et al. 2020
	Acid hydrolyzed starch			YM 50.0–69.0 MPa TS 3.1–3.2 MPa EB 63.0–80.0% TT 1.6–2.0 MJ m <sup>-3</sup> Th 300.0 μm			
Cassava (18% amylose, 82% amylopectin)	Native	Rosemary extract, glycerol, and water	Extrusion and compression	YM 9.3–23.3 MPa TS 1.8–2.3 MPa EB 49.0–70.0% TT 65.0–92.0 kJ m <sup>-3</sup> Th 300.0 μm	WVP 9.4–17.6 x 10 <sup>-7</sup> g m <sup>-1</sup> h <sup>-1</sup> Pa <sup>-1</sup>	AA 29.5–140.9 μmol TE g <sup>-1</sup>	Estevéz-Areco et al. 2010
Cassava (18% amylose, 82% amylopectin)	Native	Lentil protein isolate, glycerol, and water	Bench casting	YM 4.1–12.0 MPa TS 0.8–1.2 MPa EB 58.0–93.0% TT 4.4–5.3 x 10 <sup>6</sup> J m <sup>-3</sup> Th 0.3 μm	WVP 7.9–10.1 x 10 <sup>-7</sup> g m <sup>-1</sup> h <sup>-1</sup> Pa <sup>-1</sup>	-	Ochoa-Yepes et al. 2019
			Extrusion and compression	YM 15.0–22.0 MPa TS 1.5–2.4 MPa EB 38.0–55.0% TT 5.6–6.6 x 10 <sup>6</sup> J m <sup>-3</sup> Th 0.3 μm	WVP 5.0–5.4 x 10 <sup>-7</sup> g m <sup>-1</sup> h <sup>-1</sup> Pa <sup>-1</sup>	-	
Cassava	Native	Glycerol, cellulose fibers, and water	Tape-casting	YM 9.0–11.2 MPa TS 28.1–34.5 MPa EB 5.7–8.3% Th 100.0–400.0 μm	WVP 1.7–4.5 x 10 <sup>-7</sup> g m <sup>-1</sup> h <sup>-1</sup> Pa <sup>-1</sup>	-	Moraes et al. 2015

Cassava (15.6% amylose)	Native	Glycerol	Casting	YM 3.9 MPa TS 1.7 MPa EB 40.9%	WVP $5.0 \times 10^{-7}$ g $\text{m}^{-1} \text{h}^{-1} \text{Pa}^{-1}$	-
	Esterified			YM 2.3 MPa TS 3.0 MPa EB 115.9%	WVP $4.7 \times 10^{-7}$ g $\text{m}^{-1} \text{h}^{-1} \text{Pa}^{-1}$	-
	Cross-linked			YM 5.0 MPa TS 10.1 MPa EB 181.1%	WVP $4.3 \times 10^{-7}$ g $\text{m}^{-1} \text{h}^{-1} \text{Pa}^{-1}$	-
	Oxidized			YM 12.7 MPa TS 6.2 MPa EB 66.4%	WVP $5.4 \times 10^{-7}$ g $\text{m}^{-1} \text{h}^{-1} \text{Pa}^{-1}$	-
Waxy corn (0% amylose)	Native	Glycerol	Casting	YM 4.1 MPa TS 1.4 MPa EB 35.7%	WVP $8.3 \times 10^{-7}$ g $\text{m}^{-1} \text{h}^{-1} \text{Pa}^{-1}$	-
Corn (28.3% amylose)	Native			YM 6.7 MPa TS 1.8 MPa EB 26.7%	WVP $6.5 \times 10^{-7}$ g $\text{m}^{-1} \text{h}^{-1} \text{Pa}^{-1}$	-
Potato (21.5% amylose)	Native			YM 7.7 MPa TS 2.8 MPa EB 51.7%	WVP $6.5 \times 10^{-7}$ g $\text{m}^{-1} \text{h}^{-1} \text{Pa}^{-1}$	-
Sweet potato (16.8% amylose)	Native			YM 5.0 MPa TS 0.9 MPa EB 13.5%	WVP $6.1 \times 10^{-7}$ g $\text{m}^{-1} \text{h}^{-1} \text{Pa}^{-1}$	-
Wheat (26.9% amylose)	Native			YM 3.2 MPa TS 0.7 MPa EB 16.1%	WVP $6.5 \times 10^{-7}$ g $\text{m}^{-1} \text{h}^{-1} \text{Pa}^{-1}$	-

Dai et al. 2019

Tapioca	Native	Glycerol	Casting	YM 0.8 MPa TS 0.8 MPa EB 137.0% Th 136.5 $\mu\text{m}$	-	-	Zółek-Tryznowska et al. 2021
Rice	Native	Glycerol	Casting	YM 9.6 MPa TS 1.8 MPa EB 49.0% Th 145.1 $\mu\text{m}$	-	-	
Oat	Native	Glycerol	Casting	YM 1.8 MPa TS 0.4 MPa EB 27.0% Th 266.9 $\mu\text{m}$	-	-	
Potato	Native	Glycerol	Casting	YM 14.5 MPa TS 3.1 MPa EB 70.1% Th 332.7 $\mu\text{m}$	-	-	
Maize	Native	Glycerol	Casting	YM 14.2 MPa TS 51.0 MPa EB 1.5% Th 266.8 $\mu\text{m}$	-	-	
Cassava (26.7% amylose)	Native	Glycerol	Casting	YM 8.0–127.0 MPa TS 1.9–6.7 MPa EB 51.0–166.0% Th 60.0–210.0 $\mu\text{m}$	WVP 1.9–4.8 x $10^{-7} \text{ g m}^{-1} \text{ h}^{-1} \text{ Pa}^{-1}$	-	Luchese et al. 2018
Corn (28.5% amylose)	Native	Glycerol		YM 79.0–120.0 MPa TS 4.4–5.4 MPa EB 41.0–113.0% Th 80.0–190.0 $\mu\text{m}$		WVP 2.7–4.0 x $10^{-7} \text{ g m}^{-1} \text{ h}^{-1} \text{ Pa}^{-1}$	

Wheat (27.4% amylose)	Native	Glycerol		YM 36.0–52.0 MPa TS 2.1–2.6 MPa EB 61.0–79.0% Th 100.0–220.0 $\mu\text{m}$	WVP 3.5–4.5 x $10^{-7} \text{ g m}^{-1} \text{ h}^{-1} \text{ Pa}^{-1}$	-	
Corn	Native	Glycerol and corn starch nanocrystals	Reactive extrusion and compression	YM 0.009–0.013 MPa TS 1.1–1.2 MPa EB 76.0–282.0% TT 0.5–2.0 x $10^5 \text{ J m}^{-3}$ Th 125.0–130.0 $\mu\text{m}$	WA 3.9–5.8%	-	Gutiérrez and Valencia 2021
Corn	Crosslinker			YM 0.04–0.09 MPa TS 1.0–2.2 MPa EB 61.0–188.0% TT 0.5–3.0 x $10^5 \text{ J m}^{-3}$ Th 128.0–130.0 $\mu\text{m}$	WA 4.6–5.4%		
Corn (24.6% amylose)	Native	Fructose and corn husk fiber	Casting	YM 60.0–639.6 MPa TS 6.8–12.8 MPa Th 195.0–265.0 $\mu\text{m}$	-	-	Ibrahim et al. 2010
Corn	High amylose starch	Glycerol and water	Extrusion followed by casting	Ps 12.5 N EB 9.3 mm	WVP 9.7 x $10^{-8} \text{ g}$ $\text{m}^{-1} \text{ h}^{-1} \text{ Pa}^{-1}$	-	Calderón- Castro et al. 2018
Maize (25% amylose, 75% amylopectin)	Native	Gelatin type A (crosslinker), glycerol or sorbitol, and water	Bench-casting	TS 51.6–117.0 MPa EB 2.9–6.9% Th 30.0–70.0 $\mu\text{m}$	WVP 1.1–2.5 x $10^{-4} \text{ g m}^{-1} \text{ h}^{-1} \text{ Pa}^{-1}$		Fakhouri et al. 2013
			Pressing	TS 13.1 MPa EB 2.4% Th 40.0 $\mu\text{m}$	WVP 2.4 x $10^{-4} \text{ g}$ $\text{m}^{-1} \text{ h}^{-1} \text{ Pa}^{-1}$	-	
			Extrusion and blowing	Th 0.3 mm TS 22.6 MPa EB 1.7 %	WVP 4.4 x $10^{-4} \text{ g}$ $\text{m}^{-1} \text{ h}^{-1} \text{ Pa}^{-1}$		

Rice	Native	Glycerol	Casting	TS 2.5–3.5 MPa EB 64.3–11.4% Th 158.0–159.0 $\mu\text{m}$	-	-	Colussi et al. 2017
	Acetylation (high amylose)			TS 0.9–3.2 MPa EB 95.9–106.0% Th 148.0–158.0 $\mu\text{m}$	WVP 2.9–4.0 x $10^{-7} \text{ g m}^{-1} \text{ h}^{-1} \text{ Pa}^{-1}$	-	
	Acetylation (medium amylose)			TS 1.1–2.2 MPa EB 82.7–109.1% Th 149.0–156.0 $\mu\text{m}$	WVP 2.2–4.3 x $10^{-7} \text{ g m}^{-1} \text{ h}^{-1} \text{ Pa}^{-1}$	-	
Potato	Native	Glycerol	Casting	YM 45.1 MPa TS 3.9 MPa EB 81.0% Th 77.1 $\mu\text{m}$	WVP 8.7 x $10^{-7} \text{ g m}^{-1} \text{ h}^{-1} \text{ Pa}^{-1}$	-	La Fuente et al. 2020
	Ozone			YM 61.1–64.1 MPa TS 3.3–4.2 MPa EB 19.2–28.4% Th 61.0–64.7 $\mu\text{m}$	WVP 9.4–10.6 $10^{-7} \text{ g m}^{-1} \text{ h}^{-1} \text{ Pa}^{-1}$	-	
Yam	Native	Glycerol	Casting	TS 2.2–2.9 MPa EB 13.0–16.6% Th 33.0–60.0 $\mu\text{m}$	WVP 2.4–4.6 x $10^{-6} \text{ g m}^{-1} \text{ h}^{-1} \text{ Pa}^{-1}$	-	Narváez-Gómez et al. 2021
Yam	Oxidized			TS 4.2–5.6 MPa EB 9.9–13.3% Th 28.0–62.0 $\mu\text{m}$	WVP 1.6–3.7 x $10^{-6} \text{ g m}^{-1} \text{ h}^{-1} \text{ Pa}^{-1}$	-	
Yam	Cross-linked			TS 5.4–6.1 MPa EB 8.5–12.4% Th 35.0–98.0 $\mu\text{m}$	WVP 1.9–5.4 x $10^{-6} \text{ g m}^{-1} \text{ h}^{-1} \text{ Pa}^{-1}$	-	
Yam	Oxidized/cross-linked			TS 4.5–6.0 MPa EB 9.1–12.7% Th 33.0–67.0 $\mu\text{m}$	WVP 3.1–4.9 x $10^{-6} \text{ g m}^{-1} \text{ h}^{-1} \text{ Pa}^{-1}$	-	

Pearl millet (18.2% amylose content)	Native			Th 105.0 $\mu\text{m}$				Bangar et al. 2021
Pearl Millet (14.9% amylose content)	Heat moisture	Glycerol	Casting	Th 101.0 $\mu\text{m}$	-	-		
Pearl Millet (14.3% amylose content)	Microwave			Th 99.0 $\mu\text{m}$				
Pearl Millet (19.5% amylose content)	Sonication			Th 104.0 $\mu\text{m}$				
Jackfruit seed	Native	Anthocyanin (from black grapes) and glycerol	Casting	TS 4.4-8.2 MPa EB 13.3-45.1% Th 58.0-98.0 $\mu\text{m}$	WVP 2.3–3.5 x $10^{-6} \text{ g m}^{-1} \text{ h}^{-1} \text{ Pa}^{-1}$	A small color change was observed in the developed films when applied as fish and shrimp packaging.		Costa et al. 2021
Banana	Native	Glycerol	Casting	YM 5.1 MPa TS 0.9 MPa EB 90.3%	-	-		Silvia et al. 2021
Banana	Chemical			YM 1.7–8.9 MPa TS 0.3–0.5 MPa EB 56.5–98.3%	-	-		

Apple	Native	Glycerol and ellagic acid	Casting	YM 1.8–4.6 MPa TS 6.5–9.6 MPa EB 52.4–65.1% Th 102.8–104.4 $\mu\text{m}$	WVP 2.3–2.7 x $10^{-7} \text{ g m}^{-1} \text{ h}^{-1} \text{ Pa}^{-1}$	The increase in ellagic acid in the films significantly impacted the increase in the antioxidant capacity of the films.	Tirado-Gallegos et al. 2018
Not reported (12.5% moisture, 0.3% lipids, 0.2% proteins, 22.5% amylose)	Hydroxypropyl (0.03 degree of substitution)	Glycerol and water	Extrusion blowing	TS 5.3–9.9 MPa EB 35.0–157.6% Th 196.0–394.0 $\mu\text{m}$	WVP 4.4–0.6 x $10^{-5} \text{ g m}^{-1} \text{ h}^{-1} \text{ Pa}^{-1}$	-	Gao et al. 2021

Legend: “-” (information not available), YM (Young’s modulus), TS (tensile strength), EB (elongation at break), TT (tensile toughness), Th (thickness), Ps (puncture strength), WVP (water vapor permeability), WA (water absorption), MC (moisture content), and AA (antioxidant activity).

### **CAPÍTULO III**

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## **Cassava starch films for food packaging: trends over the last decade and future research**

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## **ABSTRACT**

Cassava starch is one of the most available and cost-effective biopolymers. This work aimed to apply a bibliometric methodology to identify the most impactful scientific data on cassava starch and its residues for food packaging in the last ten years. As a result, an increasing interest in this subject has been observed, mainly in the past five years. Among the 85 selected scientific publications, Brazil and China have been leading the research on starch-based films, accounting for 39% of the total. The International Journal of Biological Macromolecules was the main scientific source of information. Besides cassava starch, 41.18% of these studies added other biopolymers, 5.88% added synthetic polymers, and 4.71% added a combination of both. Studies analyzed suggested that different modifications in starch can improve films' mechanical and barrier properties. In addition, 52.94% of articles evaluated the film's bioactivity. Still, only 37.65% assessed the performance of those films as food packaging, suggesting that more studies should be conducted on assessing the potential of these alternative packages. Future research should consider scale-up methods for film production, including cost analysis, assessment life cycle, and the impact on the safety and quality of a broader range of foods.

**Keywords:** Biodegradable film; Agro-industrial waste; Biopolymers

The synthetic plastic crisis has led to an increasing interest in the development of biodegradable materials, particularly in developing countries, which has consequently impacted in the number of campaigns requesting the removal of single-use plastics packaging [1], [2], [3]. The replacement of petroleum-based polymers with sustainable-alternatives is, currently, technically possible for most conventional plastic materials. Moving towards the circular economy system, bio-based films, as the next generation of polymeric materials, have been considered a viable alternative to reduce greenhouse gas emissions [4].

The European Commission stated bioplastics as a leading market in the growth of the new bioeconomy [5], [6]. Most of these biopolymers are made from carbohydrate-rich plants, known as food crops, which are also currently referred to as the “first generation feedstock” of bioplastics [7], [8].

Starch is the most promising raw material to produce biodegradable polymers due to its accessibility, abundance, and low cost. Corn is the main source of starch produced in the world (65 %), followed by sweet potato starch (13 %) and cassava starch (11 %) [9], [10], [11]. Starch is a semi-crystalline polymer naturally produced in granule form by certain plants, such as roots, tubers, and cereals, in which it plays the function of an energy reserve [12]. Its composition and form vary according to the botanical source, as well as the genetic varieties of each species; starches are nevertheless composed of two polymers: amylopectin, which is the main polymer in starch, and has  $\alpha$ -1,4-linked glucose chains (degree of polymerisation (DP) < 100) joined by  $\alpha$ -1,6-linked branch points, and amylose, that constitutes < 35 % of starches and is composed of long  $\alpha$ -1,4-linked linear chains (typically DP 100–10,000) with rare  $\alpha$ -1,6-linked branch points [10], [13].

Cassava (*Manihot esculenta*) is a crop with high industrial value in several regions around the world, including South America, Asia, and Africa, and alongside corn is considered one of the main sources of starch. Cassava starch (known as “tapioca”) production is increasing rapidly (over 3 % annually), and currently represents around 7 % of the total starch produced globally [14], [15].

The use of cassava starch for the production of bioplastics has been studied extensively [16], [17], [18]. Starch-based packaging materials present some notable benefits: biodegradability, renewable source (sustainable production), good oxygen barrier ability and stretchability, as well as suitable transparency, odor and taste; they also present a lower retrogradation rate compared to other types of packaging, resulting in more stable materials over time [19]. Thus, cassava starch can be considered a key material in terms of facilitating economic growth within the new technological market trends.

However, the industrial processing of cassava to obtain starch generates a high amount of organic matter [20]. According to Pandey et al. [21], every 1 ton of cassava crop processed results in approximately 10.6 m<sup>3</sup> of liquid waste (known as cassava wastewater) with approximately 1 % solids, plus 6.4 kg of solid peel and approximately 1.1 tons of fibrous biomass residue, known as cassava bagasse. Cassava bagasse has a high moisture content, approximately [22], [23].

Thus, there is a clear need to develop and standardize methodologies to evaluate the use, types of processing and applicability of cassava and/or its residues with potential as food packaging. This study aimed (a) to investigate the profile of papers published from 2012 to 2022 on cassava starch films with potential use for food packaging through the methodology of bibliometric analysis, (b) to analyze the main characteristics of the developed films (such as mechanical and barrier properties, and bioactivity) and their impact when applied as food packaging, and (c) to briefly discuss the use of cassava starch as a precursor of food packaging materials in the context of sustainability.

## 2. Methods

### 2.1 Bibliometric analysis

The analysis of the reports on starch-based films from cassava and/or its residues with potential applicability as food packaging was carried out using the bibliometric review method [24]. The descriptors were selected to define five main areas: (i) type of material developed; (ii) possible characteristics and designations of the material developed; (iii) applicability of the material developed as food packaging; (iv) source of starch (i.e., cassava starch obtained by raw material or residues); and (v) starch. The data used for this study were extracted from two databases (Scopus and Web of Science) with the following descriptors: (“Film” OR “Bioplastic” OR “Plastic” OR “Biopolymer” OR “Composite” OR “Nanocomposite” OR “Nanobiocomposite” OR “Biocomposite” OR “Polymeric”) AND (“Biodegradable” OR “Sustainable” OR “Green” OR “Edible” OR “Active” OR “Intelligent” OR “Smart” OR “Antibacterial” OR “Antimicrobial” OR “Antioxidant” OR “Bioactive” OR “pH-sensitive”) AND (“Food packag\*”) AND (“Cassava” OR “Manipueira” OR “Cassava wastewater” OR “*Manihot Esculenta*”) AND (“Starch”).

The review of the title, abstract and keywords was conducted to determine the relevance of the papers to be selected for further bibliometric analysis. The review process was carried out on July 27, 2022. Fig. 1 shows a flow chart of the process of identification, screening, and

inclusion of articles in this review. The inclusion criteria were as follows: research paper published in journal with impact factor; full text available in English, paid and/or open access; published in the period from January 1, 2012 to July 27, 2022. Studies that did not meet the inclusion criteria were eliminated. Articles that met the inclusion criteria were organized in a spreadsheet to facilitate data management.

## 2.2 Qualitative analysis of selected articles

A literature review was carried out using a qualitative approach, considering the papers selected in the bibliometric analysis (see Section 2.1). The selected papers were grouped into sub-themes (shown in Fig. 2) to facilitate the analysis and further discussion about the main characteristics of the developed films and the impact of their application as food packaging. The sub-themes were created based on the formulation of the films, considering the studies that developed films incorporating other components, in addition to cassava starch, such as: fruit or vegetables, nanoparticles, essential oil, vegetable oil, other biopolymers, isolated bioactive compounds, synthetic polymers and/or their mixture. About 81.18 % of the 85 papers selected in Section 2.1 were included in these sub-themes ( $n = 69$ ). The remaining papers ( $n = 16$ ), despite not being grouped into sub-themes, were also considered in this discussion.

## 3. Results and discussion

### 3.1 Bibliometric analysis and scientific performance

#### 3.1.1 *Database and quantitative analysis*

There is a clear increasing trend on the scientific production of cassava starch-based films from 2012. In 2012, only two research papers on the potential of cassava starch as raw material for bioplastic production were published, both of which were from Brazilian universities (one from the Federal University of Bahia in collaboration with the Federal University of Minas Gerais and the other one from the Federal University of Santa Catarina). In the subsequent years, up until 2018, yearly production of research papers published remained constant, with a maximum in 2017 of 7 publications. Albeit moderate, the growth in the number of research studies using cassava starch to replace conventional synthetic packaging can be explained by the establishment of the 17 Sustainable Development Goals (SDGs) as an integral part of the United Nations (UN) 2030 Agenda, adopted in late 2015 [25].

Since 2019, the number of publications has increased sharply, with the latter three years of our target time study occupying 59 % of the total publications collected in the present study

(85). The largest number of publications were found in 2021, with 21 scientific reports, and in 2020, with 17 scientific reports. The period of intensified increase in the studies on this field is contemporaneous with the outbreak of the Covid-19 pandemic, during which the population were made aware of the hygienic-sanitary potential of food packaging for the protection and preservation of its content [26], [27].

In total, on the Scopus databases the papers were cited 2451 times, with an average of 28.84 citations per paper and on the WOS 1835 times, with an average of 21.59 citations per paper. The collaboration index (CI), which was calculated as the total number of authors of multiple-authored articles divided by the total number of multiple-authored articles [28], is 5.16. Only one paper published in 2020 by Kannat [29] is a single-authored paper.

### 3.1.2 *Keywords Analysis*

To present the main subjects studied in the 85 published research papers in the last decade Fig. 3 depicts the co-occurrence of the most used author's keywords in which each circle represents a keyword and its size indicates the number of publications that have the corresponding term in their keywords created with the help of flourish.studio (Canva UK Operations Ltd UK company).

The largest gray, purple, green, and blue circles refer to the most common keywords among the collected data, namely: “food packaging”, “cassava starch”, “starch”, and “active packaging” that appeared 20, 14, 13 and 10 times, respectively, and represent the most explored subjects in the field. An analysis of the data shows that other keywords are frequently used, such as “mechanical properties”, “antioxidant”, and “antimicrobial”. These research topics received consistent attention over the last ten years which can be explained by their importance in food systems whether for storage, conservation or even for extending the shelf life of packaged foods, thus are considered desirable characteristics in food packaging [30], [31], [32].

### 3.1.3 *Countries of Publications*

Based on the authors' nationality of the 85 articles published, Fig. 4 shows the global scientific production of five different continents (Africa, Asia, Europe, North America, and South America), bringing the total number of countries that have studied the topic to 27. More specifically, Fig. 5 presents the relationship among the most productive countries, journals that have published the most on the subject and the years in which these papers were published on a Sankey diagram.

Single Country Publications consider scientific papers from a one country alone. By analyzing the obtained data, this review found 65 Single Country Publications. Brazil boasts the highest scientific contribution in the field of cassava starch-based films, with a total of 25 Single Country Publications, followed by China with 10 Single Country Publications. The large number of publications of Brazilian institutions can be linked to the high accessibility and widespread cultivation of cassava crops in this country, which motivate national researchers [33].

On the other hand, Multiple Country Publications demonstrate the interaction between authors of different nationalities and realities. In this study, at least one Multiple Country Publications per country was considered, and as a result we observed a network with 20 countries. Brazil and The United States had the highest association index with the same association number, 5 Multiple Country Publications for each.

#### *3.1.4 Publications and the main 5 journals*

To analyze the scientific impact of the research developed with cassava starch-based films in the last ten years, Table 1 identifies the 5 most relevant journals based on the number of publications on the topic. Regarding the journals with the same number of publications, the highest impact factor (data obtained from the journal's official website in August 2022) was used as a choice criterion. In short, the main 5 journals identified were: 1. International Journal of Biological Macromolecules, with 12 published papers (impact factor of 8.025 in August 2022); 2. Carbohydrate Polymers, with 7 published papers (impact factor of 10.723 in August 2022); 3. Food Hydrocolloids, with 6 published papers (impact factor of 11.504 in August 2022); 4. Food Packaging and Shelf Life, with 6 published papers (impact factor of 8.749 in August 2022); and 5. Polymers, with 4 published papers (impact factor of 4.967 in August 2022).

The total number of articles published in the above-mentioned journals was 35, among them 26 Single Country Publications and 8 Multiple Country Publications. Brazil was the country with the most relevant contribution with 11 scientific reports, followed by China with 8 and Thailand with 8 scientific reports.

#### *3.1.5 Worldwide publications and cassava crop production*

As has already been discussed, cassava crop has largely been used in recent years for animal or human consumption, either for direct consumption or for industrial processing, from which different food products are obtained, such as flour and starch [34]. In addition, only a

small portion (10 %) of its application is destined to the textile, pulp, and paper industries [35], that is, there is massive potential to apply the crop as a biopolymer source [16], [18]. As a result of its ability to easily adapt to different environmental conditions as well as its high commercial value, cassava crops are cultivated in >100 countries, and is one of the most produced tuberous roots in the world [36].

To highlight this, Fig. 6 presents the relationship between scientific production of bio-based films using cassava starch and the global production of cassava from 2012 to 2020, the year of the latest available data according to the FAO database [36]. In 2012, even with high global production, the number of publications using cassava for the development of biodegradable plastics was relatively low. However, it is possible to observe a peak in publications in 2020, the first year in which global cassava production reached 300 million tons. This data corroborates the fact that cassava starch-based polymers can be considered a new commercial trend that is driving the increase in demand and consequently the production of the cassava crop globally.

### 3.2 Cassava starch films as food packaging

There are numerous advantages in using cassava starch as a film base such as good film-forming capacity, transparency, biodegradability, and non-toxicity [19]. However, to make this material more competitive and suitable for application in the food packaging sector, some studies focus on strategies that provide greater mechanical and barrier resistance to the films, such as the use of modified starch and incorporation of other components in the film-forming solution, such as plasticizers (glycerol and sorbitol), nanoparticles, and other biopolymers and synthetic polymers [32], [37], [38], [39]. Thermoplastic starch (TPS) has been widely used as a base for films or in mixtures with other polymers since the plasticization of starch favors the improvement of the mechanical and barrier properties of the films [16]. Dang & Yoksan [40] evaluated the impact of the plasticizer used in the film-forming formulation on the mechanical and barrier properties of the film. It was observed that the molecular size of the plasticizers influenced the properties of the films. The film added with glycerol showed lower tensile strength and water vapor barrier property, compared to films added with larger plasticizers, such as xylitol and sorbitol [40].

Regarding starch modifications, they can be classified as chemical, physical, enzymatical, and their combinations [38]. The most common is chemical modification, which introduces a functional group to the starch molecule by derivation or decomposition reactions, and physical modification, which modifies the starch structure by thermal or non-thermal

means [39]. Some of the selected reports on the bibliometric analysis investigated the impact of using modified starch compared to native starch to formulate films. Limin Dai et al. [41] observed that modified starch films (esterified, cross-linked, and oxidized cassava starch) showed greater transparency and better mechanical properties with an average increase of 274% in TS, 196 % in EB, and 75% in YM when compared to the native cassava starch film. Among the modified starches, the use of oxidized cassava starch and adipic acid cross-linked cassava starch resulted, respectively, in films with greater rigidity (higher YM value, 12.65 MPa), mechanical strength (TS of 10.12 MPa), and elasticity (EB of 181.07%). On the other hand, acetate esterified cassava starch generated a film with higher TS (3.02 MPa) and EB (115.91%), but lower YM (2.66 MPa) than the film with native cassava starch (1.72 MPa of TS, 40.92% of EB, and 3.87 MPa of YM). Regarding water vapor permeability (WVP), only adipic acid cross-linked cassava starch resulted in a film with greater water barrier properties than native starch, probably due to the replacement of hydroxyl groups with ester groups that are hydrophobic [41]. Similarly, Torrenegra et al. [42] modified three different starches, including cassava, through hydrolysis, esterification and acetylation reactions. Compared to native starch films, starches modified using docecynyl succinic anhydride made the films more rigid and resistant, while starches modified with octenyl succinic anhydrid and acetic anhydride generated films with greater elasticity and flexibility [42]. In another study, the use of modified cassava starch in the film-forming solution generated films with lower WVP ( $0.92$  to  $1.92 \times 10^{-10} \text{ g m}^{-1} \text{ s}^{-1} \text{ Pa}^{-1}$ ) and EB values (47.65 to 92.01 %) and higher TS values (6.41 to 13.65 MPa) than films with native cassava starch ( $2.13 \times 10^{-10} \text{ g m}^{-1} \text{ s}^{-1} \text{ Pa}^{-1}$  WVP; 102.44% EB; and 5.73 MPa TS). Furthermore, modified cassava starch films presented better transparency and water-resistance than native cassava starch films [43]. The results of these studies suggested that different modifications in starch can contribute to the improvement of the mechanical and barrier properties of the films since, when modifying the chemical structure of the native starch molecule, the chemical bonds can become stronger, favoring the formation of a more structurally integrated polymer matrix.

In addition to starch modifications, polymer blending can also contribute to improving the properties of the starch film. Among the 85 selected papers in the bibliometric analysis, 41.18% (n = 35) developed films with the addition of other biopolymers, 5.88% (n = 5) with the addition of synthetic polymers, and 4.71% (n = 4) with the addition of both, in addition to cassava starch (Fig. 2). One example is the study by Ferreira et al. [44] that evaluated the impact of different mixtures between cassava starch and soy protein isolate on film properties. In general, the films with the highest protein concentration were more rigid (YM of 4.4 MPa),

more soluble in water (35.52%) and had better barrier properties (WVP of  $1.56 \text{ g mm kPa}^{-1} \text{ h}^{-1} \text{ m}^{-2}$ ), probably due to the higher crystallinity of these films and crosslinking of the polymeric chain. On the other hand, as the proportion of starch in the mixture increased, the intermolecular interactions between starch and protein reduced the crystalline areas of the films, making the polymer network more mobile (EB of 84.98, 39.23, 21.29, 14.33, and 8.27% for the 100:0, 85:15, 70:30, 55:45, and 40:60 starch:protein films, respectively) [44]. Similarly, starch films with pea protein showed higher TS and lower flexibility, probably due to the strong molecular interaction of the biopolymer mixture, as well as the inadequate matrix homogeneity, which presented protein aggregation. Moreover, the higher the concentration of protein in the mixture, the higher the WS, WVP, oxygen permeability, and hydrophobicity of the surface of the films [45]. Castro et al. [46] evaluated the properties of starch and gelatin-based films. The higher gelatin concentration in the film promoted greater flexibility (98.98 % increase in EB), thermal reinforcement and barrier property (19.73% reduction in WVP) when compared to the formulation with neat cassava starch. However, the film with only starch showed higher mechanical strength (YM of 1116 MPa and TS of 14.6 MPa) than the other formulations with different concentrations of gelatin/starch. After optimizing the formulation, the authors concluded that the best starch:gelatin ratio was 53:47, resulting in the best mechanical and water barrier properties, particularly for application as food packaging [46]. In another study, De Lima Barizão et al. [47] observed that the addition of  $\kappa$ -carrageenan to cassava starch and PVA films reduced their flexibility, with an EB value varying from 4.36 % to 67.65 %, for films with high to low concentration of  $\kappa$ -carrageenan, respectively. Furthermore, the higher proportion of  $\kappa$ -carrageenan increased the mechanical strength and stiffness of the films (TS of 25.88 MPa and 59.64 MPa of YM). Nevertheless, a higher concentration of starch in the mixture was associated with higher WS values in the films, increasing from 39.22 % (film with 1:0  $\kappa$ -carrageenan:starch) to 62.86 % (film with 0:1  $\kappa$ -carrageenan:starch) and higher thermal stability, suggesting that carrageenan may have disrupted the semicrystalline structure of starch in the polymer matrix [47]. Mangaraj et al. [17] observed that the higher concentration of starch mixed with PLA resulted in films with lower strength (TS of 25.16 MPa), oxygen transmission rate ( $123.92 \text{ cc m}^{-2} \text{ day}^{-1}$ ) and greater flexibility (EB of 5.49%), probably due to interruptions in the polymer matrix. Despite the negative impact of starch on some properties of the films, the authors concluded that all formulations (with higher or lower concentrations of starch) were suitable for application for modified atmosphere packaging of capsicum, even when compared to the non-biodegradable polymers tested [17].

Nanoparticles can also be used in the formulation of starch films to reinforce the polymer matrix and improve its mechanical, barrier, and thermal properties, since they have a higher aspect ratio than their larger-scale equivalents. In general, nanoparticles interfere with the crystalline and molecular network of the matrix, affecting the movement of the polymer chain [32], [37], [48]. In this bibliometric analysis, 23.53% (n = 20) of the articles added nanoparticles, with an improvement being observed in the mechanical, thermal and/or barrier properties of the films developed in most of these studies (n = 18). Menezes et al. [31] investigated the use of TiO<sub>2</sub> nanoparticles on the mechanical properties, water interaction, and optical characteristics of chitosan-cassava starch films. The TiO<sub>2</sub> nanoparticle improved the mechanical properties through electrostatic interactions and hydrogen bonds, where the film with 1% TiO<sub>2</sub> had a 15% increase in TS and 100% in EB compared to the film without nanoparticles. In addition, 1% of TiO<sub>2</sub> reduced water adsorption, probably due to its interaction with the other hydrophilic groups of the polymer matrix, which reduces the number of interactions available to water molecules, making the film less hygroscopic and more resistant to water [31]. Corroboratively, Phothisarattana et al. [49] also observed that the homogeneous dispersion of TiO<sub>2</sub> nanoparticles increased mechanical strength and reduced permeability to oxygen, carbon dioxide and water vapor in nanocomposite films of poly(Butylene adipate-co-terephthalate) and thermoplastic starch. Cellulose nanocrystals were also incorporated into cassava starch film, improving the physicochemical properties of the films. In another study, Ma et al. [50] observed that carboxymethyl cellulose nanocrystals from sweet potato residue improves the physicochemical properties of cassava starch-based nanocomposite films, showing an increase of 554% in TS, 41% in EB and 123% in WS and a reduction of 42.78% in WVP and 15.9% in moisture absorption compared to the cassava starch film with the incorporation of cellulose nanocrystals.

Nanoparticles can often provide antimicrobial potential to starch films. Shapi'i et al. [32] developed a cassava starch film with the chitosan nanoparticle and observed antimicrobial activity (by disc diffusion analysis) especially against Gram-positive bacteria (*Bacillus cereus* and *Staphylococcus aureus*), in addition to improved shelf life of cherry tomatoes with the inhibition of microbial growth (when compared to food packaged in neat starch film). Srikhao et al. [51] evaluated the effect of oregano essential oil and silver nanoparticles in polyvinyl alcohol/starch film. The authors concluded that there was a synergistic effect on the antibacterial activity between the nanoparticle and the essential oil and that a minimum inhibitory concentration of 5% of the essential oil was sufficient for antimicrobial activity against *Escherichia coli* and *Staphylococcus aureus*. This formulation

also favored obtaining a film with greater efficiency in scavenging the DPPH radical (88.42%) when compared to a film with only essential oil (59.54%) or only nanoparticle (50.91%). In addition to bioactivity, the film with the addition of 5% essential oil and nanoparticle showed an increase of 80.9% in TS and 88.6% in YM compared to the film without the addition of essential oil, demonstrating greater mechanical resistance [51]. In this context, J. F. Mendes et al. [52] observed that the addition of lemongrass essential oil in starch films also presented improved mechanical properties, thermal stability, barrier to moisture, and colorimetric attributes compared to films without essential oil.

Fruit and vegetables can also be used to change the main properties in starch films and may contribute to some type of bioactivity in these films, due to the presence of antioxidant and/or antimicrobial compounds, favoring the use of the film as an active packaging for food. In this study, 27.06% of the papers (n = 23) developed starch films incorporating fruit, vegetables and/or agro-industrial residues in different forms (such as powder, flour, extract, and puree) in the film-forming solutions. P. S. Müller et al. [53] observed that the incorporation of rosemary and green tea extracts in cassava starch films with PBAT had a positive effect on the barrier property of the films, with lower WVP values. Cunha et al. [54] evidenced that the incorporation of propolis extract in starch films promoted greater extensibility in addition to providing an important content of phenolic compounds and antioxidant activity, demonstrating a potential for use as active food packaging. Another study evaluated the use of propolis by-product in cassava starch film as a way of adding value to an agro-industrial residue and increasing the properties in starch film. The addition of propolis to the film promoted greater stiffness (10.95% increase in YM) compared to the control film and was able to generate films with excellent antioxidant and antibacterial activity against *Staphylococcus aureus* and *Salmonella Typhimurium* [55].

Food packaging should primarily facilitate the transport and marketing of food, and protect it from environmental factors such as oxygen, humidity, light, and microbiological contamination [26]. Thus, for starch films to be used as food packaging, they must have a minimally high TS and YM, and a reasonable EB, to prevent breakage during handling. Other properties that are fundamental in films are barrier properties, with preferably low WVP and OP to protect food against deterioration, oxidation and dehydration, in addition to optical, sensory, toxicity, and biodegradability or compostability properties [38]. Among the articles selected in this study, 37.65% (n = 32 papers) evaluated the impact of the starch-based film developed as packaging for different foods, such as oil, fruit and vegetables, bread and biscuits, cheese and butter, yogurt, meat products, and chocolate (Fig. 7). In general, to evaluate the

performance of the films the studies presented lipid oxidation, microbial growth, general appearance, soluble solids and titratable acidity, color, light transmission, mass loss, and sensory evaluation analyzes. There are promising data on the application of starch-based films as food packaging, for instance, Kanatt [29] developed an active film of starch and gelatin with lemon juice capable of increasing the shelf life of minced chicken meat for up to 12 days under refrigeration, minimizing oxidative changes. Corroboratively, Lin et al. [30] also observed greater protection against lipid peroxidation in chicken meat packed in edible films of cassava starch, carboxymethyl cellulose and apple polyphenol compared to the film-formulation without polyphenol. In this sense, Shapi'i et al. [32] observed an increase in the shelf life of cherry tomatoes packed with starch films and chitosan nanoparticles compared to cherry tomatoes wrapped using neat starch film and, especially, unwrapped, being the only samples that showed microbial growth above the acceptable level after 10 days of refrigerated storage. The positive effects observed in these films were associated with plant extracts and/or bioactive compounds incorporated in filmogenic formulations, suggesting that starch-based films are compatible with different antioxidant/antimicrobial components and may favor the development of active food packaging.

In recent years, interest in research and development in active packaging has grown. This type of food packaging includes characteristics such as antimicrobial, antioxidant, and aromatic activity. Active packaging, unlike conventional packaging, interacts with food, improving its nutritional, sensory, chemical and/or microbiological quality. The bioactive compounds incorporated and/or immobilized in the polymeric matrix of the packaging can migrate to the food product, maintaining or prolonging its shelf life [26], [48]. Another possibility is the development of smart or intelligent packaging, in which a certain compound is incorporated into the film with the function of monitoring any change in the food, including pH, or the environmental condition, such as relative humidity, signaling the quality status of the food product from manufacturer to consumer [48], [56].

Through the bibliometric analysis of this research a total of 45 papers (52.94 %) were found that evaluated the bioactivity of the films through analyses of the antioxidant and antimicrobial properties and/or through the impact on the quality of the food packaged in the film (application as active packaging). Among the studies that evaluated the bioactivity directly in the packaged food, Friedrich et al. [57] developed a starch and gelatin-based coating containing *Tetradenia riparia* extract with the objective of delaying the ripening process of strawberries stored under refrigerated conditions. In fact, despite the extract having increased thickness, opacity and WVP, the authors obtained films with properties suitable to be applied

as packaging for strawberries, acting efficiently during the 10 days of cold storage, controlling bacterial growth and preserving the high antioxidant activity of the fruit. Lin et al. [30] obtained cassava/sodium carboxymethyl cellulose film with apple polyphenols with an increase in the barrier property (reduction of WVP values) and greater thermal stability compared to the control film (without bioactive compound), probably due to the high compatibility of the formulation components (creation of hydrogen bonds), improving the crystallinity and cohesion of the matrix. As expected, the film with apple polyphenols showed antioxidant properties, proving to be suitable for preserving chickens, inhibiting their oxidation and increasing shelf life [30].

Among the studies selected in the bibliometric analysis, some evaluated the potential of the developed film to be considered smart packaging. Cheng et al. [43] developed smart cassava starch films added to red cabbage extract that favored a broad and intense colorimetric response detectable to the naked eye in the pH range 2 to 12 and with some color reversibility. The anthocyanins present in the extract and responsible for the characteristic color were properly stabilized in the film by electrostatic interactions and hydrogen bonds with the starch [43]. Yun et al. [58] also developed starch films with antioxidant and pH-sensitive properties using anthocyanin-rich bayberry extract. The lower extract concentration contributed to better water vapor barrier properties and mechanical strength, due to a dense and compact internal microstructure, unlike the higher extract concentrations that agglomerated in the film, generating a more heterogeneous matrix. In addition, the starch films with extract showed important antioxidant activity and were able to monitor the freshness of pork by evaluating sensitivity to pH variation [58]. Another study showed that the incorporation of green tea and basil extracts into starch films also showed potential as active and smart films, since they showed high antioxidant activity, due to high phenolic content, and color dependency on pH variation, due to the presence of chlorophyll and carotenoids in the extracts. Other positive aspects of the incorporation of the extracts were rapid degradation in soil (less than two weeks) and thermostability up to 240°C, in addition to lower WVP and maintenance of flexibility compared to the control film [59]. Therefore, considering the aforementioned data, cassava starch-based films can be considered promising for wide applications such as active and smart packaging for different food systems.

It is important to emphasize that the COVID-19 pandemic may have contributed to a greater awareness of the importance of packaging for the protection and conservation of food, increasing food safety and reducing waste, as well as the need to use new alternatives of

materials that are more sustainable [26]. Item 3.2 addresses the use of cassava starch as a food packaging material in the context of sustainability.

### 3.3 Cassava starch films: a sustainable alternative for food packaging material?

In recent years, due to increased environmental awareness and stricter environmental regulations, mainly considering single-use plastics, there has been a growing interest in finding new, more sustainable materials for food packaging, with a particular interest in biodegradable materials [26], [48]. The study's bibliometric analysis highlighted that about 17.65% of the papers ( $n = 15$ ) analyzed the biodegradability in films (Table 2).

From the studies presented in Table 2, it is possible to observe that the incorporation of different reinforcement components in starch-based films did not significantly affect the biodegradability of the material, maintaining high rates of biodegradation. There are different methods to evaluate the biodegradability of polymers. Further research on the best biodegradation conditions in different systems and for each polymer is necessary to understand the real impact of these materials after disposal. However, it is important to point out that the biodegradability of the films alone is not necessarily associated with the greater sustainability of these films, compared to conventional polymers. A more holistic assessment of the environmental impact is required, such as greenhouse gas emissions and water and land use throughout the life cycle, from raw material extraction to final disposal [60]. Finally, other factors must be considered, such as the economic impact of the polymer and, when applicable, the costs associated with adequate environmental recovery. These issues must be investigated so that decisions and changes have scientific support. For example, De Léis et al. [61] investigated the energy and environmental performance of producing cassava starch-based film in Brazil using Life Cycle Assessment as a tool. The authors concluded from the analysis of different hypothetical scenarios that the energy and environmental impacts come from the cultivation of cassava and the consumption of electrical energy to produce the additives used in the formulation of the film and the film itself. In general, industrial processes had a greater impact than agricultural processes. Furthermore, the authors observed that the use of a mixture of cassava starch and polyethylene with the addition of renewable glycerin in the film formulation and production by extrusion appeared to have the lowest energy impact but had the worst environmental performance regarding terrestrial acidification due to the polyethylene processing [61]. Another recent study found that despite the higher CO<sub>2</sub> emission of cellulose and starch biopolymers compared to some synthetic bioplastics (polyvinyl chloride, polypropylene, polyethylene, and polyethylene terephthalate), these biopolymers are

superior in terms of the reduction in land and water use during production [5]. Overall, bioplastics tend to reduce non-renewable energy use and greenhouse gas emissions compared to conventional plastics [5].

Another relevant factor is the use of agro-industrial residues to extract biopolymers and other compounds of interest for film formulation. The analysis of bibliometric data showed that only 2 articles (2.35%) extracted starch from cassava residues and 14 articles (16.47%) incorporated another residue directly and/or some component extracted from vegetable residues (e.g., peel/ husk, leaves, and straw). Some authors point out that using starch, an important energy source for human nutrition, could be diverted from the food industry to the development of biomaterials [62]. Thus, it reinforces the importance of focusing our efforts on extracting biopolymers from waste that would otherwise be discarded, thus presenting a more adequate alternative for use and better waste management.

#### **4. Conclusions and Perspective**

The bibliometric analysis showed that the first half of the last 10 years was marked by an irrelevant number of publications when compared to the world scientific trends, with an average of 3 publications per year. However, after the establishment of the 17 Sustainable Goals of the United Nations 2030 Agenda, bioplastics were described as leading the new bioeconomy with rapid a growth in the number of publications over the past 5 years. In terms of the number and distribution of publications by country, Brazilian and Chinese researchers stood out, and together were responsible for 39% of the published papers.

Regarding the performance of cassava starch as a biomaterial, the results of these studies suggest that chemical structure modifications to the native starch are needed to form stronger bonds, favoring the formation of a more structurally integrated polymer matrix. In addition to starch modifications, blending starch with other polymers is an alternative that can also contribute to production of biodegradable improved films. It is worth mentioning that only two papers used starch from cassava residues. Considering the importance of cassava starch for human nutrition, further research on extracting starch from cassava waste, both solid (such as peel and bagasse) and liquid (cassava wastewater) is recommended. Such research should include more adequate alternatives for use, thus contributing to waste management, the generation of sustainable products with high added value and in turn reducing environmental impacts.

Additional research that focuses on new technologies for starch-based films, especially on an industrial scale, is necessary to improve the properties and expand the application of these

films as packaging for various foods, such as fruits and vegetables, bakery products and meat. Research gaps which could be explored include the search for cost reduction strategies, the increase of production on an industrial scale (rather than laboratory scale) optimizing the filmogenic formulation and process conditions, the evaluation of the entire life cycle of films, in addition to investigating the impact on the safety and quality of packaged foods, also aiming to reduce waste.

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### **Author contributions**

J. R. V. Matheus contributed to the conceptualization, collected the data, gathered and interpreted background literature, contributed to data analysis and writing the manuscript; P. M. Farias gathered and interpreted the background literature, contributed to data analysis and writing the manuscript; J. M. Satoriva contributed to the collection and analysis of data; C. J. Andrade contributed to the critical revision of the final version of the manuscript; A. E. C. Fai contributed to the conceptualization and the critical revision of the final version of the manuscript, provided funding, and supervised the project.

### **Conflicts of interest**

There are none to declare.

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**Table 1.** Five most relevant journals and outcomes of interest of the published papers.

Ranking	Journal/ Impact Factor*	Year	Outcomes of Interest	Country	Reference
1	International Journal of Biological Macromolecules (8.025)	2019	<ul style="list-style-type: none"> <li>- Different starch-based films are fully characterized.</li> <li>- Starches from different plant sources for film formation are studied.</li> <li>- Amylose content has a major effect on the physicochemical properties of the film.</li> <li>- Cross-linked starch film is the most suitable for food preservation.</li> <li>- The results can assist in the matrix selection of starch-based film.</li> </ul>	China	[40]
		2019	<ul style="list-style-type: none"> <li>- Lycium ruthenicum anthocyanins (LRA) were added to starch film.</li> <li>- LRA addition improved the barrier ability and tensile strength of starch film.</li> <li>- LRA could interact with starch chains through hydrogen bonds.</li> <li>- Starch-LRA films showed antioxidant and intelligent pH-sensitive properties.</li> <li>- Starch-LRA films could be used to monitor the freshness of pork.</li> </ul>	China	[63]
		2020	<ul style="list-style-type: none"> <li>- Cassava starch:kappa-carrageenan:glycerol:poly(vinyl alcohol) blends provide films.</li> <li>- A low water vapor permeability occurs at 50:50 starch:kappa-carrageenan molar ratio.</li> <li>- The thermal stability increases with the addition of starch in the blend.</li> <li>- The kappa-carrageenan is responsible for producing stiff films.</li> <li>- The starch supports films with high elongation at break.</li> </ul>	Brazil	[46]
		2020	<ul style="list-style-type: none"> <li>- The composite film prepared show good mechanical and hydrophobic properties and is a candidate for food packaging.</li> <li>- Stearic acid can be used to modify microcrystalline and nanocellulose successfully.</li> <li>- Both the M-MCC and M-NCC can enhance the mechanical and hydrophobic properties of the film.</li> </ul>	China	[64]
		2020	<ul style="list-style-type: none"> <li>- Efficiency of a coating based on native cassava starch (NCS), gelatin, and sorbitol, containing different concentrations of <i>Tetradenia riparia</i> extract.</li> <li>- The addition of extract increased the thickness, opacity, and water vapor transmission rate (WVTR), but decreased the solubility of the films when compared to the control.</li> <li>- The extract incorporation improved the control over bacterial growth and preserved the high antioxidant activity of the strawberries within ten days of storage.</li> </ul>	Brazil	[57]
		2020	<ul style="list-style-type: none"> <li>- Novel antibacterial and biodegradable starch composite films were proposed.</li> </ul>	China	[65]

	<ul style="list-style-type: none"> <li>- Mechanical properties and water vapor barrier of our films were reinforced with WMSNs/KC.</li> <li>- For the first time, starch-based films with SA achieved antimicrobial activity.</li> <li>- A significant bacteriostatic effect was obtained in yogurt in contact with our films.</li> <li>- This product was nontoxic, edible and could extend the shelf life of food.</li> </ul>		
2021	<ul style="list-style-type: none"> <li>- Gelatinized starch is a residue from the starch processing classification step.</li> <li>- Gradual substitution of the cassava starch by gelatinized starch was conducted.</li> <li>- The visual aspect was modified as a result of the increase in the residue content.</li> <li>- Biodegradable starch-based films were manufactured by the casting technique.</li> <li>- It was possible to produce samples containing only residual starch.</li> </ul>	Brazil	[62]
2021	<ul style="list-style-type: none"> <li>- Intermolecular interactions and crystal structures of the polymer matrix were not strongly affected by the addition of LAE.</li> <li>- Elongation at break of blend films were significantly improved by the synergistic effect of acetic acid, glycerol, and LAE.</li> <li>- Active films with only 1% LAE possessed antibacterial ability.</li> </ul>	China	[66]
2021	<ul style="list-style-type: none"> <li>- Blends of native and crosslinked cassava starch were prepared by solution casting.</li> <li>- 10 wt% kaolin improved water resistance and tensile strength of the blend film.</li> <li>- Sachets from a starch blend film with 10 wt% kaolin were a promising application.</li> </ul>	Thailand	[67]
2021	<ul style="list-style-type: none"> <li>- Nopal cladode flour, propolis extract and lignin were added to cassava starch film.</li> <li>- Nopal cladode was subject to alkaline treatment (NC12) to produce films (S-NC12).</li> <li>- S-NC12 were less hydrophilic, less permeable to water vapor and more flexible than S-NC.</li> <li>- Films with NC had a greater phenolic compounds content and antioxidant activity than lignin and propolis.</li> </ul>	Brazil	[17]
2021	<ul style="list-style-type: none"> <li>- Influence of the plasticizer type (glycerol, glycerol/xylitol, and glycerol/sorbitol) and content on the performance of TPS blown films.</li> <li>- TPS mixed with glycerol, a small-sized plasticizer, was easily processed and extensible, however the surface stickiness lead to single-wall films, low tensile strength, and poor water vapor barrier properties</li> <li>- When replacing glycerol for larger-sized plasticizers such as xylitol or sorbitol, the films exhibited reduced stickiness and separable double walls and showed improved mechanical and barrier properties.</li> </ul>	Thailand	[68]
2022	<ul style="list-style-type: none"> <li>- Cassava starch (CS) and sodium carboxymethyl cellulose (CMC) edible films as basic raw materials and apple polyphenol (AP) as biologically active components.</li> <li>- Addition of AP to the film increased its barrier performance and thermal stability.</li> </ul>	China	[30]

			- The application of CS/CMC/AP film in chicken preservation proved its antioxidant potential.		
2	Carbohydrate Polymers (10.723)	2014	- Development of biodegradable films impregnated with cinnamaldehyde. - Supercritical solvent impregnation (SSI) was used to produce active films. - Total amount of impregnated cinnamaldehyde depends on CO <sub>2</sub> solubility. - Water vapor permeability of the films decreased after SSI.	Brazil; Portugal	[69]
		2016	- Biodegradable and edible cassava starch films with yerba mate extract were developed. - TGA, DSC, ATR/FTIR and DRX tests demonstrated plasticizing effects of the extract. - Extract compounds of low molecular weight reduced starch–starch chain interactions. - The extract used limited crystal growth, recrystallization, and starch retrogradation. - The degradation in compost of films with yerba mate extract was faster than matrix.	Argentina; Venezuela	[70]
		2016	- The X-ray patterns and TEM images confirm the good dispersion of silicate layers in the LLDPE/TPS blend. - Introduction of organoclay has enhanced the thermal decomposition of nanocomposite materials. - The obvious improvements in tensile properties and storage modulus of DMA were observed on the nanocomposite samples. - The nanocomposite film at high TPS content showed more than 60% of mass loss after 5 months in compost soil.	Viet Nam; France	[71]
		2017	- Active and smart biodegradable films from starch and natural extracts were developed. - The films with green tea and basil extracts showed high antioxidant activity. - Films changed color at different pH, resulting in good indicators of food quality. - Starch-natural extract showed faster biodegradability and high thermal degradation.	Argentina	[59]
		2019	- The addition of CMC increased mechanical properties of starch films. - Corn starch film presented hydrophobic properties due to high amylose content. - The presence of CMC in starch film reduced water vapor permeability.	Brazil	[72]
		2020	- Two lactic acid bacteria (LAB) strains with a high exopolysaccharide yield were selected from pickled water. - Edible films based on cassava starch were developed containing LAB and sodium carboxymethylcellulose (CMC). - The antioxidant activity of the composite films was significantly enhanced. - Lactobacillus plantarum showed uniform distribution in the cassava starch/CMC matrix. - The composite films can extend the shelf life of banana.	China; USA	[73]
		2020	- Antimicrobial activity of starch/CNP films is dependent on the concentration of CNP.	Malaysia	[32]

			<ul style="list-style-type: none"> <li>- 15 to 20% w/w starch/CNP films can inhibit bacterial growth.</li> <li>- CNP inhibit gram-positive bacteria efficiently compared to gram-negative bacteria.</li> <li>- Shelf life of cherry tomatoes wrapped in starch/CNP films improved.</li> </ul>		
3	Food Hydrocolloids (11.504)	2014	<ul style="list-style-type: none"> <li>- Films were prepared from cassava starch, cashew tree gum (CTG) and carnauba wax (CW).</li> <li>- The hydrophobic CW improved water vapor barrier and water resistance of films.</li> <li>- CW impaired the film transparency because it formed a non-miscible phase.</li> <li>- The relative proportions of cassava starch and CTG did not affect film properties.</li> </ul>	Brazil	[74]
		2015	<ul style="list-style-type: none"> <li>- Edible films based on cush-cush yam and cassava starches were developed.</li> <li>- The structure of cush-cush yam film showed a lower gelatinization than the cassava starch.</li> <li>- Barrier and mechanical properties suggested strong glycerol–starch interaction.</li> <li>- In modified films, especially of cassava, glycerol–starch interaction was stronger.</li> </ul>	Venezuela; Argentina	[75]
		2017	<ul style="list-style-type: none"> <li>- Rosemary extracts (RE) were successfully incorporated in cassava starch films.</li> <li>- Active films showed significant antioxidant activity.</li> <li>- UV-properties of the films were enhanced due to the RE presence.</li> <li>- RE incorporation inhibited the bonding between glycerol and starch molecules.</li> <li>- Films containing RE showed high biodegradation after 14-days of composting.</li> </ul>	Argentina; Colombia	[76]
		2020	<ul style="list-style-type: none"> <li>- Emulsions were obtained by the combination of Tween 80/pectin and lemongrass oil (LEO).</li> <li>- Active films of TPS/pectin/LEO were scaled up through a continuous casting approach.</li> <li>- The emulsified films showed improved mechanical properties.</li> <li>- The TPS/pectin/LEO films showed complete biodegradation in vegetal compost.</li> </ul>	Brazil	[52]
		2021	<ul style="list-style-type: none"> <li>- Bilayer films of starch, with gellan or xanthan, and polyesters were obtained by thermoprocessing.</li> <li>- Gellan and xanthan gums improved the mechanical properties of starch films.</li> <li>- Both gums decreased water vapour and oxygen permeability of starch films.</li> <li>- Starch-polyester bilayers presented high barrier capacity to water vapour and oxygen.</li> <li>- Bilayers with cassava starch-gellan showed the best properties for food packaging.</li> </ul>	Spain	[19]
		2022	<ul style="list-style-type: none"> <li>- Intelligent films were made using red cabbage extracts (RCEs) and dual-modified starch.</li> <li>- The dual-modified starch films exhibited high water-resistance and good tensile strength.</li> <li>- RCEs were fixed into films by electrostatic interactions and hydrogen bonding.</li> </ul>	China	[42]

4	Food Packaging and Shelf Life (8.749)	2019	<ul style="list-style-type: none"> <li>- Demonstrate potential industrial process of modified starch as active packaging.</li> <li>- Extruded LLDPE with 30 and 40% TPS-GT films showing active functions.</li> <li>- Release of phenolics and antioxidants were controlled by microstructures.</li> <li>- Process–structure-property relationships of LLDPE/TPS films in blown extrusion.</li> <li>- Green tea incorporated blown films preserved meat color and quality of packaged oil.</li> </ul>	Thailand; France	[77]
		2020	<ul style="list-style-type: none"> <li>- Active packaging from conventional blown extrusion of herb compounding TPS/LLDPE.</li> <li>- Films with sappan and cinnamon provided antimicrobial properties and preserved redness of beef.</li> <li>- Cinnamon provided higher compatible nano- and micro-structures between starch and LLDPE.</li> </ul>	Thailand	[78]
		2020	<ul style="list-style-type: none"> <li>- Pea protein prevented stickiness and improved processability of blown-extruded starch.</li> <li>- Pea protein enhanced barrier properties of starch films.</li> <li>- Addition of pea protein into starch acetate at up to 20 % formed no clumps.</li> <li>- Protein improved oxidative stability and structural integrity of edible package.</li> </ul>	Thailand	[44]
		2020	<ul style="list-style-type: none"> <li>- Active films present high barrier to UV light and antioxidant properties.</li> <li>- Rheological properties of the FFS were not affected by the additives.</li> <li>- Structural heterogeneities due to Natamycin caused brittleness in active films.</li> <li>- Cassava starch/chitosan active films with Natamycin present antifungal activity.</li> </ul>	Italy; Brazil	[79]
		2022	<ul style="list-style-type: none"> <li>- Functionalized edible films with papain improved meat tenderness within 30 min.</li> <li>- Interaction between starch and papain involved hydrogen bonding.</li> <li>- Film elasticity is controlled by enzyme concentrations and starch types.</li> <li>- Papain reduced water sensitivity and stabilized starch film structures in water.</li> <li>- Papain incorporated films improved cherry-red color of packaged beef.</li> </ul>	Thailand	[80]
		2022	<ul style="list-style-type: none"> <li>- Acetate starch gave the best mechanical strength to the PBAT and TPS blend films.</li> <li>- Hydrophobic OS starch improved compatibility and interaction with PBAT.</li> <li>- Morphology of blown PBAT/TPS controlled film properties more than miscibility.</li> <li>- Increased hydrophilicity and water absorption of starch improved biodegradation.</li> <li>- Modified TPS altered aromatic arrangement in PBAT, involving C-H and C-O bonding.</li> </ul>	Thailand	[81]
5	Polymers (4.967)	2020	<ul style="list-style-type: none"> <li>- Development of active cassava starch and carboxymethyl cellulose based films with the addition of quercetin and tertiary butylhydroquinone (TBHQ) antioxidants.</li> <li>- The addition of antioxidants improved tensile strength, but reduced elongation at break of the cassava starch–CMC film.</li> <li>- Cassava starch–CMC films containing quercetin and TBHQ showed better physical properties than cassava starch–CMC (control) film.</li> </ul>	Thailand; USA.	[82]

	- The application of cassava starch–CMC film containing quercetin and TBHQ can delay both the oxidation of lard (35–70 days) and the red discoloration of pork.		
2021	<ul style="list-style-type: none"> <li>- Study of the influence of the addition of borax (crosslinking agent) on the thermal conductivity of cassava starch biopolymer.</li> <li>- The increase in the amount of borax is proportional to the increase in the glass transition temperature.</li> <li>- The highest value of the thermal conductivity is reached at a volume fraction of 1.40% of borax added. Adding more borax is counterproductive since some of the thermal bridges get interrupted.</li> </ul>	Mexico; Colombia	[83]
2021	<ul style="list-style-type: none"> <li>- Effects of titanium dioxide (TiO<sub>2</sub>) on the morphology and properties of poly (butylene adipate-co-terephthalate) (PBAT)- and thermoplastic cassava starch (TPS)-blended films.</li> <li>- Increasing TiO<sub>2</sub> raised amorphous starch content and hydrogen bonding by interacting with the TPS phase of the polymer blend.</li> <li>- The application of blended films containing 5% of TiO<sub>2</sub> on banana fruit showed slower darkening color change and an enhanced shelf-life of 12 days.</li> </ul>	Thailand	[84]
2021	<ul style="list-style-type: none"> <li>- Development of antioxidant active food packaging materials from cassava starch/gelatin composite films with quercetin and tertiary butylhydroquinone (TBHQ).</li> <li>- Increasing the content of quercetin and TBHQ increased the film solubility in water and the WVTR of composite films.</li> <li>- The blended films were found to slow down the oxidation of lard (more than 35 days) and postpone the red discoloration of pork.</li> <li>- Cassava Starch/Gelatin composite films added with quercetin and TBHQ can be used as an active food packaging since they showed potential to delay oxidation reactions in packaged products thereby enhancing product shelf-life and quality.</li> </ul>	Thailand; USA.	[85]

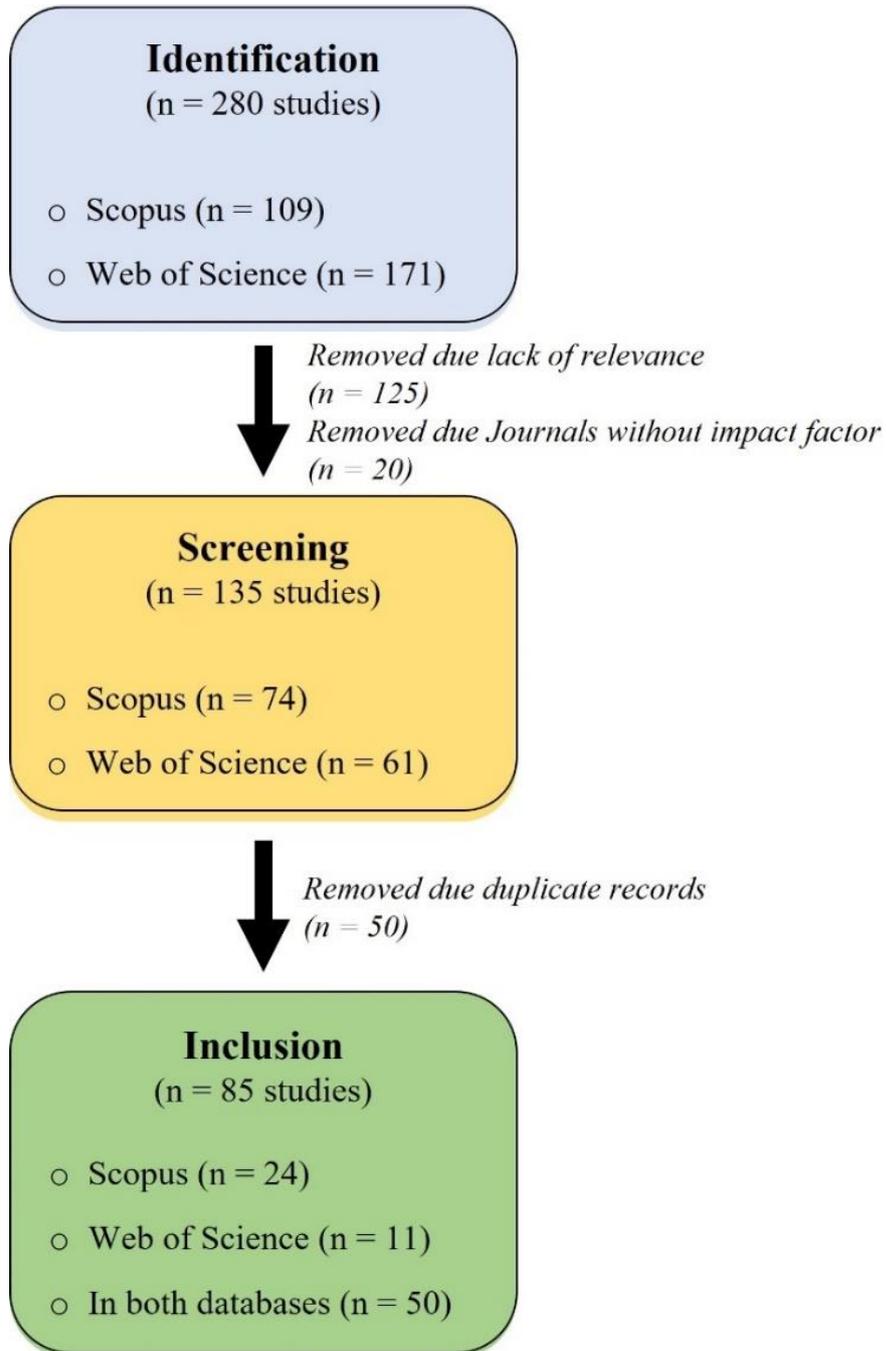
\* The impact factors were obtained from the official website of each journal in the period of August 2022.

**Table 2.** Biodegradability analysis of cassava starch-based films

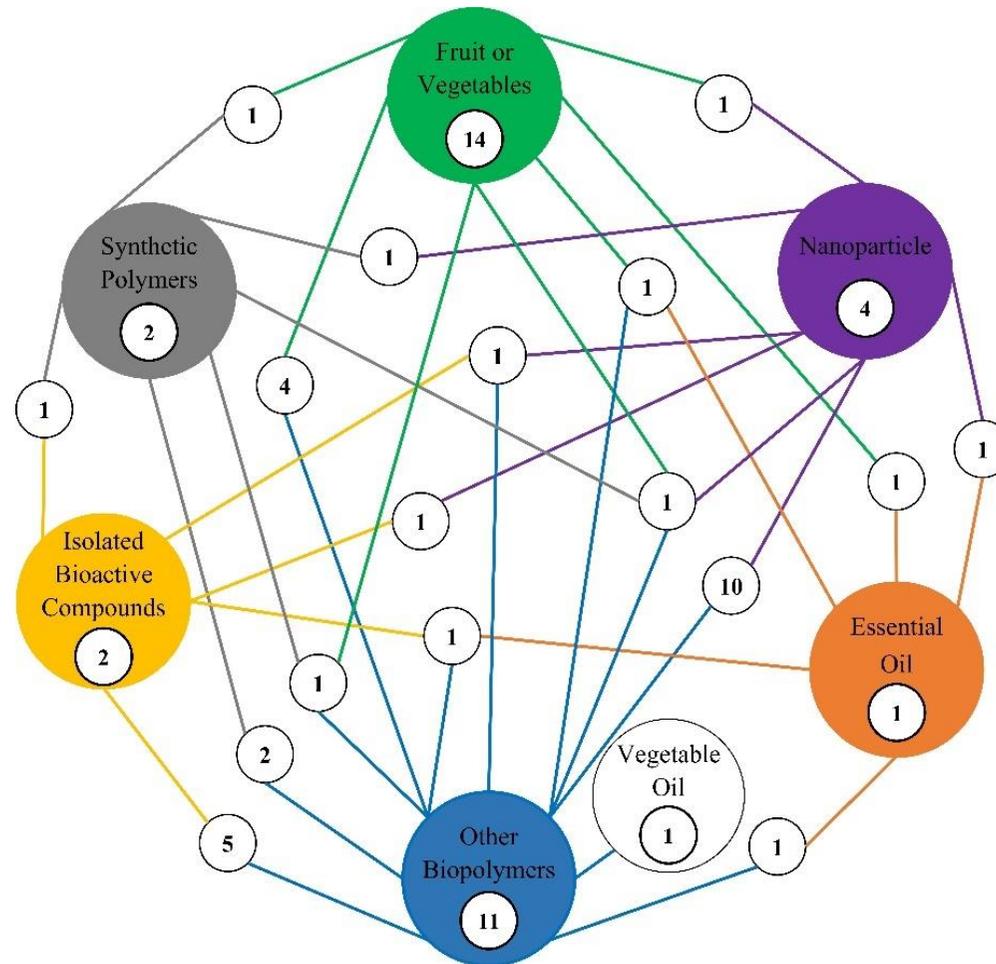
<b>Biodegradability test</b>	<b>Film formulation</b>	<b>Main results</b>	<b>Ref.</b>
Soil burial test	PLA/ Cassava starch, glycidyl methacrylate, and benzyl peroxide	- After 150 days the films showed about 47% to 52% weight loss in soil.	[18]
Soil burial test	Semi-refined ι-carrageenan, cassava starch, SiO <sub>2</sub> -ZnO nanoparticles, sodium dodecyl sulfate, and glycerol	- The developed films, probably due to antimicrobial activity, were not fully degraded after 28 days. - Control film was heavily degraded after 28 days.	[86]
Biodegradation in vegetable compost (qualitative evaluation)	Cassava starch, glycerol, and lemongrass essential oil emulsion	- The developed films showed signs of degradation on the 7th day of analysis. - The essential oil apparently did not affect the biodegradability of the starch film.	[52]
Biodegradation in vegetable compost (qualitative evaluation)	Cassava starch, glycerol, and aqueous rosemary extracts	- The films showed changes in integrity and tonality in the first 7 days. - The biodisintegration of the polymeric chains of the films may be associated with the action of heat, humidity, and enzymatic activity of microorganisms. - At the end of 14 days, films without extract were practically disintegrated, in contrast to films with extract, which were better preserved.	[76]
Biodegradation in vegetable compost (qualitative evaluation)	Cassava starch, glycerol, and green tea or basil extracts	- Films with both extracts were rapidly degraded in 6 days. - On the 12th day, the degradation was significant, suggesting that the plant extracts accelerated the degradation process in the soil.	[59]
Biodegradation in vegetable compost (qualitative evaluation)	Cassava starch, glycerol, and yerba mate extract	- All the films showed signs of degradation, with changes in tonality and breakdown. - All film formulations were practically degraded on the 12th day, especially those with the addition of extract, suggesting that this incorporation improved the biodegradability of the starch-glycerol films.	[70]
Biodegradation and composting in soil (qualitative evaluation)	Cassava starch: gelatinized starch, and glycerol	- After 135 days, it was not possible to observe fragments of the samples buried in soil with the naked eye, indicating the biodegradability of the developed films.	[62]
Biodegradation in soil	Cassava starch, glycerol, sodium carboxymethyl cellulose, <i>Lactobacillus plantarum</i> or <i>Pedococcus pentosaceus</i>	- Films without a probiotic property showed 41.14% weight loss on the 30th day, most likely caused by hydrolysis and microbial degradation. - The addition of probiotics negatively affected the degradation of the film in the soil, perhaps by reducing the more hydrophilic content in the	[73]

		formulation, reducing the swelling and WVP of the film and, consequently, absorbing less water during the degradation process.	
Biodegradation in soil (qualitative evaluation)	Thermoplastic starch, brewery spent grain, cocoa butter, and lemongrass essential oil nanoemulsions	<ul style="list-style-type: none"> <li>- The films showed changes in the tonality and integrity of the material, with significant signs of degradation at the end of the 45 days of analysis (increase in holes, breaks, and deposition of organic matter on the surface of the films).</li> <li>- Some components of the formulation, such as cocoa butter and lemongrass essential oil, may have delayed the degradation process, due to reduced permeability of the films; the films were still considered biodegradable.</li> </ul>	[87]
Biodegradation in soil (qualitative evaluation)	Cassava starch, ethyl alcohol, glycerol, and tetraethyl orthosilicate	<ul style="list-style-type: none"> <li>- The films showed rapid biodegradability in soil after 20 days, estimating by visual analysis an average degradation of about 60%.</li> <li>- The addition of tetraethyl orthosilicate apparently did not influence the biodegradability of the films.</li> </ul>	[88]
Biodegradation in soil (qualitative evaluation)	Mixture of native and modified cassava starch, kaolin, and glycerol	<ul style="list-style-type: none"> <li>- The neat starch film exhibited more erosion and a more irregular morphology than films with added kaolin at the end of 14 days.</li> <li>- The addition of kaolin reduced the degradation of the films, probably due to greater hydrophobicity in the polymer matrix, which reduces the migration of water from the soil to the films and reduces the action of microorganisms.</li> </ul>	[67]
Biodegradation in soil and in seawater	Cassava starch, chitosan, acetic acid, glycerin, and ZnO	<ul style="list-style-type: none"> <li>- On the 7th day, films without ZnO showed a 30.05% weight loss, reaching 100% of mass loss on the 28th day, unlike the films added with ZnO that showed less biodegradability in the soil, with mass loss ranging from 83.62% to 89.98% on the 28th day.</li> <li>- The addition of ZnO in the films reduced the biodegradability in soil, possibly by increasing the barrier to the water diffusion rate of the films, in addition to making them more rigid and with possible antimicrobial activity, hindering the action of microorganisms.</li> <li>- Films without ZnO were also more degradable in seawater, probably explained by the hydrophobicity of ZnO (higher ZnO content in the formulation negatively impacted the degradation).</li> <li>- On the 14th day, all samples of starch films with and without ZnO were fully decomposed in seawater. The starch, chitosan and ZnO films (at higher concentrations) were fully biodegraded in seawater on the 21st day,</li> </ul>	[89]

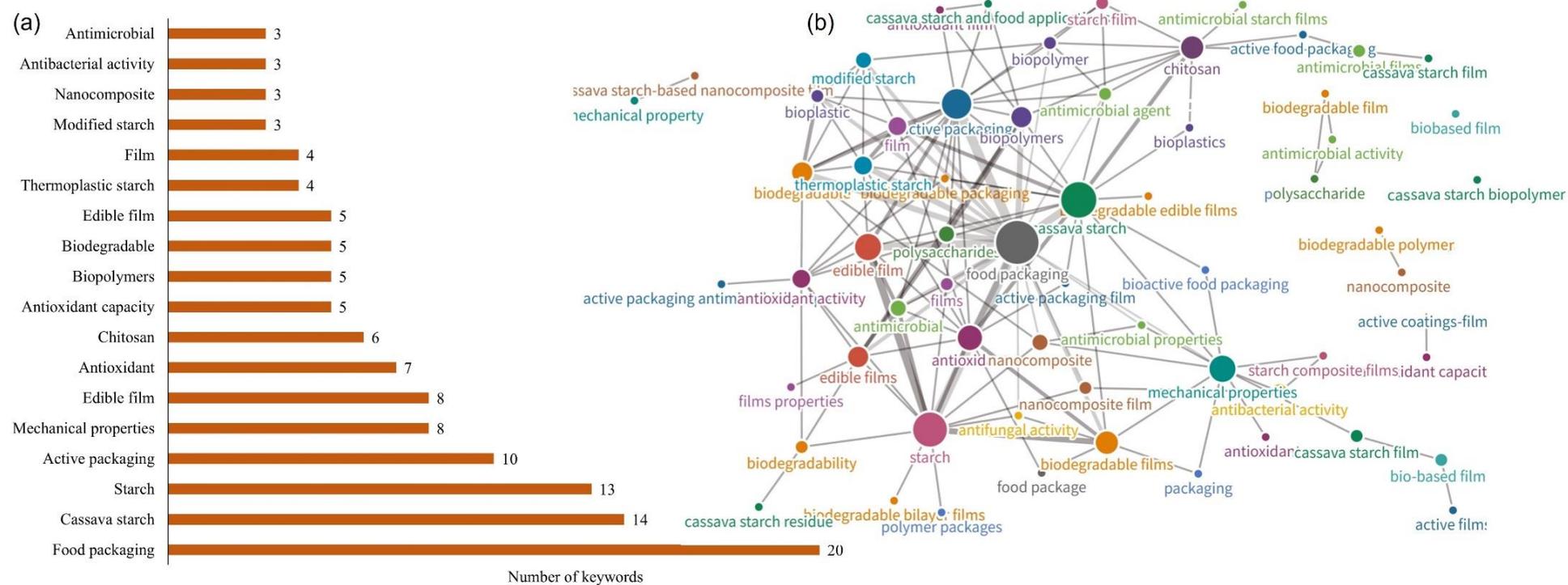
		<p>suggesting that the hydrophobicity of chitosan also had a certain negative impact on the biodegradability of the film.</p>	
Aerobic biodegradation	Native or modified cassava starch, glycerol, and polybutylene adipate terephthalate	<p>- Polybutylene adipate terephthalate and starch blend films showed a high rate of biodegradation reaching 80% biodegradation after 5–7 days of storage.</p> <p>- Different native and modified starches in polybutylene adipate terephthalate affected the rate of biodegradation (native and hydroxypropylated starch reached 99% biodegradation on day 8, acetylated starch reached 97% on day 9, and octenyl-succinated starch reached 98% biodegradation on day 11).</p> <p>- The biodegradation of the films was dependent on the hydrophilicity of the polymers, in which the native and acetylated starch generated highly hydrophilic matrices, unlike the octenyl-succinated starch which promoted the lowest hydrolytic and water activity for the film.</p>	[81]
Aerobic biodegradation	Linear low density Polyethylene (LLDPE), cassava starch, glycerol, and montmorillonite nanofil 15	<p>- The developed film showed a degree of biodegradation close to 55% in 80 days, reaching 63% of biodegradation in 150 days.</p> <p>- LLDPE/cassava starch blends seem to have favored the degradation of LLDPE, as starch is easily metabolized by a wide range of organisms, attracting microorganisms and causing a fracture in the LLDPE chain, which is not easily metabolized.</p>	[90]
Aerobic compost environment test	Native and acid hydrolyzed cassava starch and glycerol	<p>- The weight loss after 15 days of the samples was 58.3% and 45.21–47.26% for native starch and modified starch films, respectively.</p> <p>- The migration of water from the soil to the starch chain causes swelling in the films, increasing weight loss and favoring microbial action.</p> <p>- Film with native starch showed greater degradation than films with modified starches, probably due to the higher amylose sugar content.</p> <p>- The longer starch hydrolysis time during chemical modification increased the relative crystallinity of the films obtained, impacting the biodegradation rate (slight reduction in weight loss).</p>	[91]



**Figure 1.** Methodology applied to the process of identification, screening, and inclusion of articles in this review



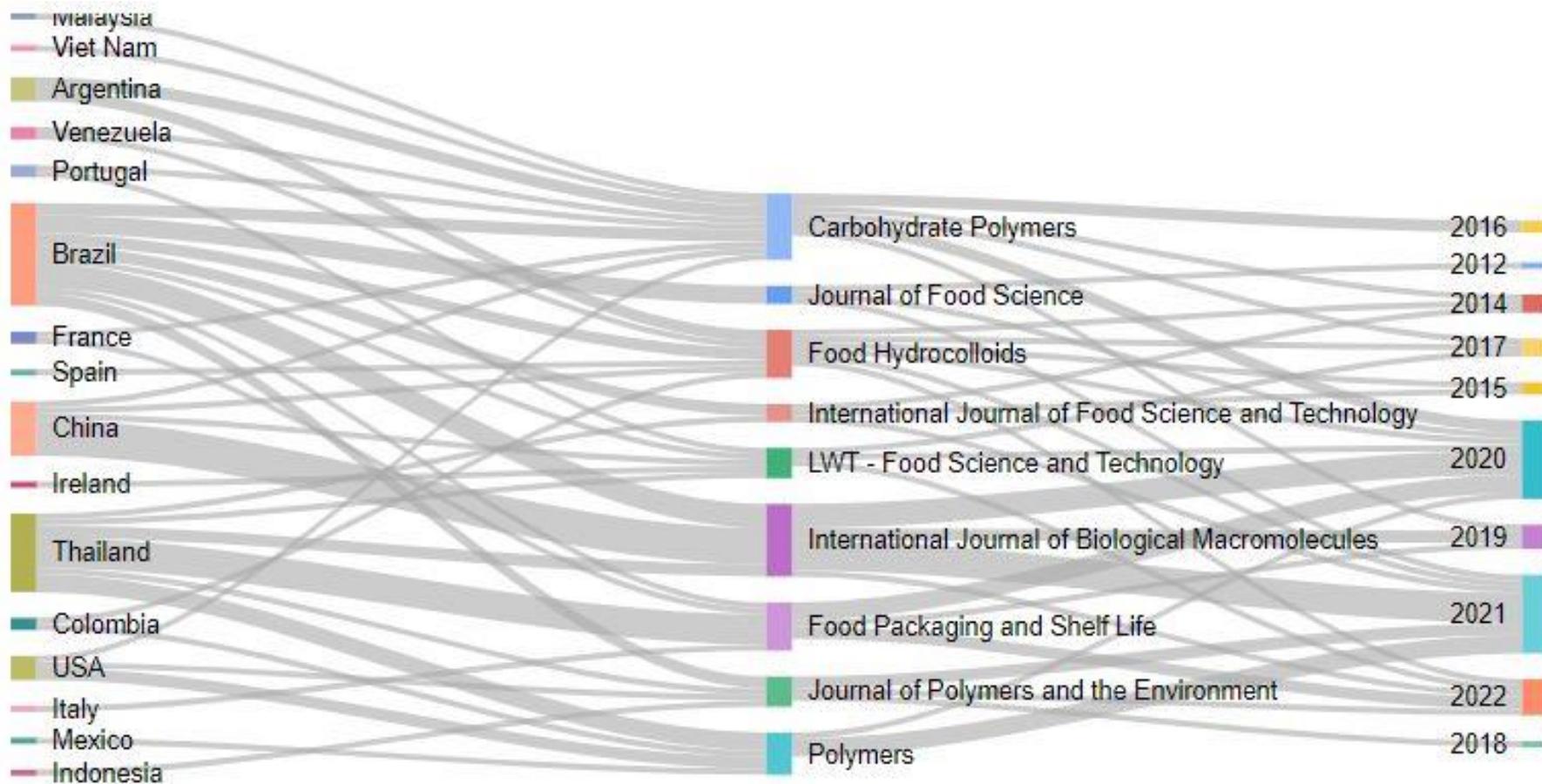
**Figure 2.** Scientific papers studying the theme related to cassava starch films from 2012 to 2022. To this end, the general theme was grouped in subthemes referring to the incorporation of several components in the filmogenic solution (fruit and vegetables, nanoparticles, essential oil, vegetable oil, other biopolymers, isolated bioactive compounds, synthetic polymers and/or the mixture of them, indicated by the crossings). The number inside of each round figure means the number of published papers with the indicated theme.



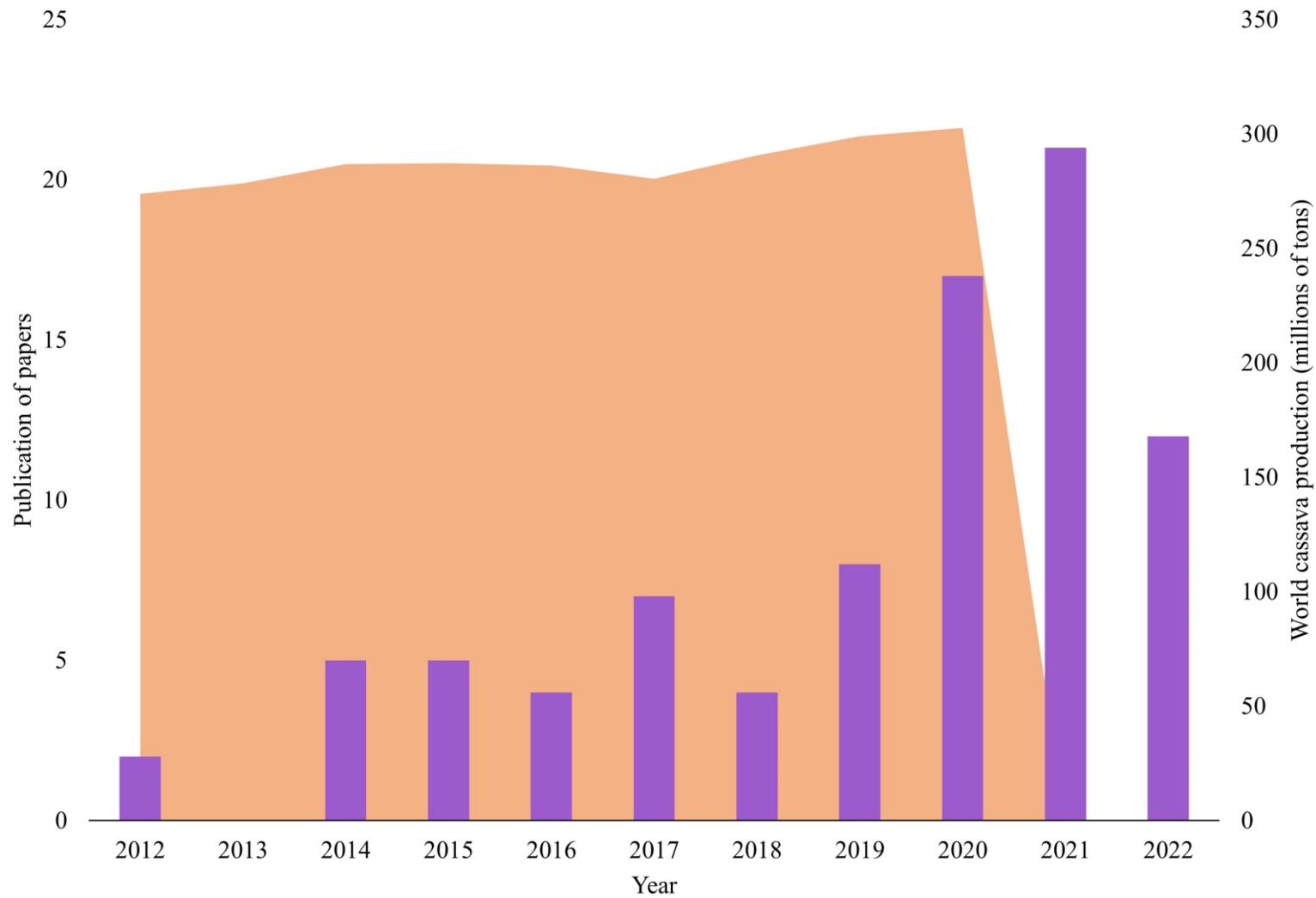
**Figure 3.** (a) Number of occurrences of the main keywords and (b) network of keywords co-occurrence present in the selected works.



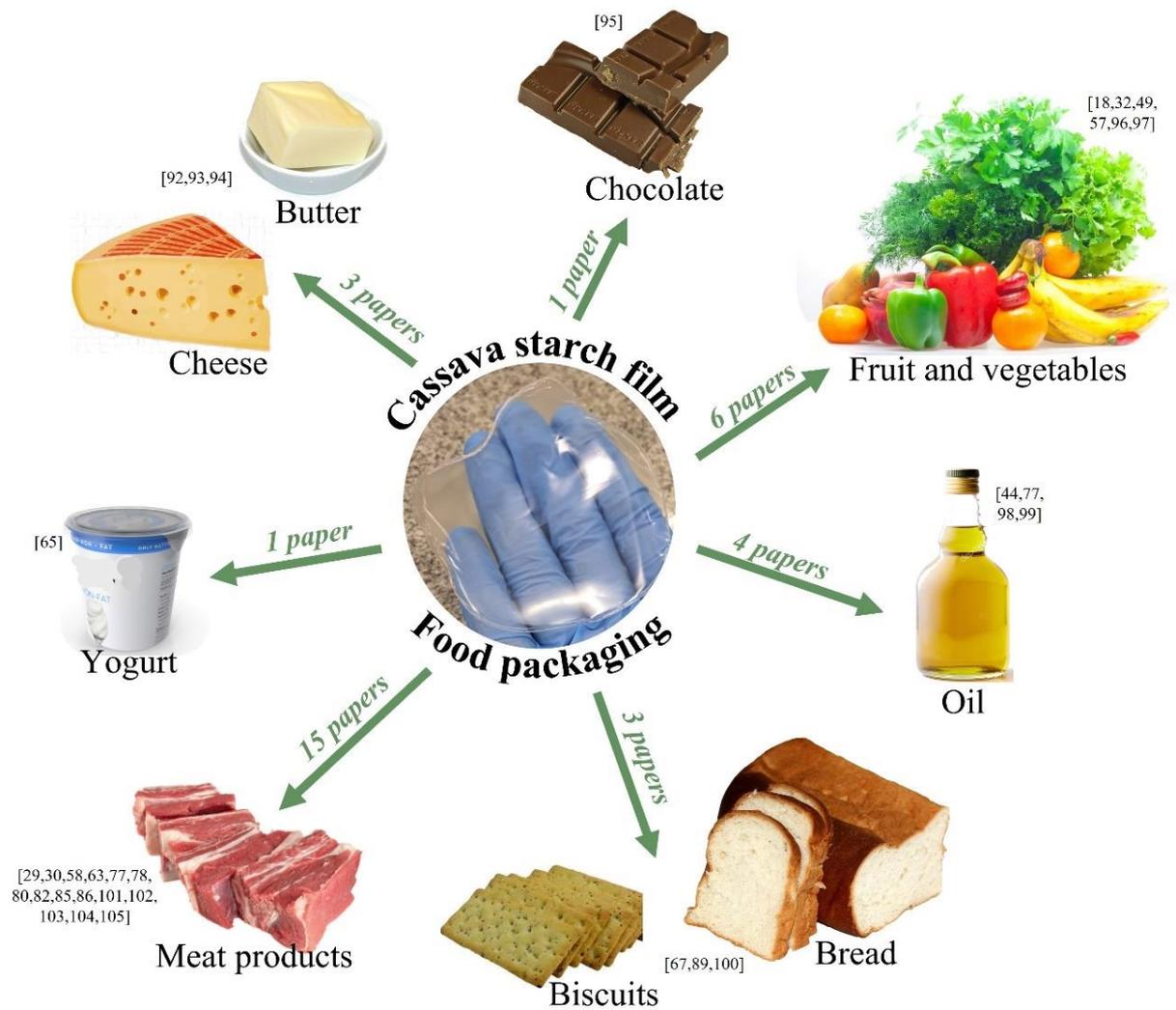
**Figure 4.** Representative map of quantitative scientific publications on cassava starch-based films for food packaging production per country between 2012 and 2022 according to the data obtained in this study.



**Figure 5.** Three-field plot of cassava starch-based films relating main journals, countries, and target years.



**Figure 6.** Correlation between the scientific production about cassava starch-based films and the world production (FAO, 2021) of cassava in millions of tons per year.



**Figure 7.** Cassava starch-based films applied as packaging for different food matrices.

## **CAPÍTULO IV**

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*Submetido – under review*

Starch-based films containing purple halochromic pigments: smart food packaging (submetido para International Journal of Biological Macromolecules).

## **CAPÍTULO V**

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*Submetido*

Closing of cycle: cassava starch and wastewater films as biostimulant for bean seed and plant growth enhancement (submetido para Biocatalysis and Agricultural Biotechnology)

## **CAPÍTULO VI**

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*Submetido*

Valorization of purple carrot peel as pH-indicating smart tag for plant-based food freshness  
(submetido para Food Hydrocolloids)

## **CAPÍTULO VII**

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*A ser submetido*

Starch/pectin materials with beetroot or purple sweet potato peels: from chemical properties to application as smart food packaging

## **CAPÍTULO VIII**

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*A ser submetido*

Olive leaves addition on starch-pectin films: optimization, characterization, and evaluation as edible hydrosoluble sachets

### 3. CONCLUSÃO

Existem muitas estratégias para aproveitar resíduos agroindustriais para a produção de bioplástico dada a diversidade química dessas matrizes e ampla gama de combinações, concentrações possíveis e formas de incorporação aos biopolímeros. Essa tese verificou a incorporação e o impacto de diferentes resíduos agroindustriais para o desenvolvimento de filmes à base de amido-pectina, para diferentes aplicações em embalagens de alimentos.

Dos resíduos utilizados se destacam as cascas de vegetais roxos e as folhas de oliveira, os quais demonstram boa aplicabilidade em formulações filmogênicas, resultando em filmes com propriedades mecânicas, físicas, físico-químicas e de barreira satisfatórias, além de capacidade de proteção contra a radiação UV, atividade antioxidante e biodegradabilidade. A caracterização dos resíduos foi fundamental para compreender o efeito da adição destes resíduos na matriz polimérica e suas implicações nas propriedades estudadas, servindo como guia para propor novas aplicações em sistemas de embalagens inteligentes e ativas de alimentos.

A composição da manipueira utilizada nesse estudo apresentou baixo teor de amido e elevado de proteína, afetando negativamente a resistência mecânica dos filmes. No entanto, como ponto positivo, destaca-se a rápida biodegradabilidade e potencial de não fitotoxicidade desses filmes, justificando esforços contínuos para melhoria da padronização da manipueira, enquanto matéria-prima. Ademais, por ser um resíduo gerado em grande volume durante todo ano no Brasil, reforça-se a importância do desenvolvimento de novas estratégias para utilizá-lo no contexto de biorrefinaria.

O resíduo de cenoura roxa apresentou alto potencial como agente pigmentador em filmes à base de amido de mandioca e pectina. Esse estudo avaliou o efeito da forma de incorporação das cascas de cenoura roxa – sob a forma de pó ou extratos. O uso do extrato aquoso de pó de casca de cenoura roxa (PCP) >100 mesh na solução filmogênica resultou em melhorias nas propriedades mecânicas. Já os filmes adicionados de pó de PCP <100 mesh ou de extrato aquoso com NADES de PCP >100 mesh foram mais responsivos, colorimetricamente, ao pH, sugerindo potencial uso como selos inteligentes. Esses filmes se mostraram eficientes quando aplicados como selos inteligentes para monitorar o frescor de um alimento plant-based análogo ao frango, com impacto positivo a segurança alimentar e ambiental. Vale destacar que não foram encontrados estudos até essa data que tenham aplicado embalagens inteligentes em produtos *plant-based* protéicos, sendo um dos aspectos inovadores desse trabalho.

A utilização de resíduos de batata doce roxa e de beterraba como matéria-prima para incorporação em formulações filmogênicas também se demonstrou adequada para obtenção de

filmes biodegradáveis, não fitotóxicos, com relevante propriedade halocrômica – indispensável para aplicação como selo colorimétrico sensível ao pH. A incorporação de ambos os resíduos em filmes à base de amido-pectina sob a forma de pó trouxe um aspecto de inovação para esse estudo, uma vez que comumente utiliza-se os resíduos sob a forma de extrato. O pó de resíduos reduziu a transparência, tornou a cor mais vibrante e resultou em elevada capacidade de proteção contra radiação ultravioleta, principalmente para aquelas formulações com maior teor de pó. Os filmes adicionados de casca de batata doce roxa em pó (PSP) se destacaram pelo aumento da resistência mecânica e sensibilidade colorimétrica à variação de pH e exposição ao vapor de amônia. Já os filmes adicionados de casca de beterraba em pó (BEET) se destacaram por uma elevada propriedade de barreira ao vapor de água, capacidade antioxidante, rápida biodegradação e potencial efeito positivo na germinação de sementes de feijão. A aplicação de ambos os filmes como selo inteligente para monitorar o frescor de um alimento *plant-based* análogo ao frango foi promissora. O revestimento dos morangos usando a suspensão filmogênica incorporada com cada um desses resíduos não foi capaz de preservar a firmeza e aparência desejável dos morangos, mesmo garantindo a estabilidade microbiológica. Por outro lado, a aplicação dos filmes como tampa, substituindo o PVC, em embalagens de morangos refrigerados também foi eficiente. Foi preservada a firmeza, o aspecto visual e atendido o padrão microbiológico, em consonância com a legislação brasileira vigente, para esse alimento.

A incorporação de pós de folhas de oliveira (< 100 mesh) em filmes de amido-pectina foi otimizada utilizando planejamento experimental de processos e superfície de resposta. O filme otimizado se destacou pela elevada solubilidade em água, mantendo propriedades mecânicas e de opacidade satisfatórias, ideal para aplicação como sachê hidrossolúvel comestível. Além disso, o filme otimizado apresentou importante capacidade de proteção contra radiação ultravioleta. O filme otimizado foi aplicado como embalagem primária, enquanto sachê de temperos e de infusão de folhas de oliveira (pó > 100 mesh), e avaliado sensorialmente por provadores não treinados. Os resultados dessa análise mostraram elevada aceitação global e ótima intenção de compra. O uso desse resíduo sob a forma de pó, a otimização da formulação, a aplicação como sachê e a análise sensorial são os principais pontos inovadores desse estudo.

A valorização de resíduos agroindustriais pode contribuir para promoção da segurança alimentar (inocuidade) a partir de processos verdes e práticas mais sustentáveis que corroboram com a redução do desperdício de alimentos, com o desenvolvimento de novos materiais biodegradáveis, compostáveis e comestíveis para embalagem ativa/inteligente de alimentos. Finalmente, este estudo está alinhado com os princípios da bioeconomia circular, com a agenda 2030 e com os diferentes objetivos transversais de desenvolvimento sustentável da ONU.

Considerando os resultados e as limitações deste estudo, sugere-se para trabalhos futuros: (1) adoção de estratégias para melhorias nas propriedades mecânicas e de barreira ao vapor de água dos filmes; (2) avaliação do envelhecimento dos filmes; (3) otimização da extração e retenção de compostos bioativos dos resíduos/ subprodutos incorporados nas formulações filmogênicas; (4) avaliação da vida útil dos alimentos embalados com os filmes/revestimentos ativos; (5) avaliação do desempenho dos filmes inteligentes no monitoramento das condições de transporte e armazenamento na cadeia fria de alimentos; (6) avaliação do ciclo de vida dos filmes; (7) avaliação quantitativa ampliada dos testes de biodegradação em diferentes ecossistemas e o impacto na germinação de diferentes tipos de sementes; (8) avaliação da aceitação das embalagens ativas e inteligentes pelos consumidores; e (9) desenvolvimento de estratégias para o escalonamento produtivo e viabilização comercial das embalagens ativas e inteligentes de alimentos.

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## ANEXO I

Santos-Maragoni C, Souza TSP, Matheus JRV, Nogueira TBB, Xavier-Santos D, Miyahira RF, Antunes AEC, Fai AEC. COVID-19 pandemic sheds light on the importance of food safety practices: risks, global recommendations, and perspectives. Crit Rev Food Sci Nutr. 2022; 62(20): 5569-81. <https://doi.org/10.1080/10408398.2021.1887078>

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## COVID-19 pandemic sheds light on the importance of food safety practices: risks, global recommendations, and perspectives

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### Abstract

The outbreak of the coronavirus disease (COVID-19) is global health and humanitarian emergency. To respond effectively to this pandemic, it is mandatory to reaffirm science in its different fields of study, including the food safety area. Presently, we review food safety in times of COVID-19, exploring whether the virus can be transmitted by food or water; recommendations from regulatory agencies; perceptions of food hygiene practices during the pandemic; and post-pandemic perspectives. The review was based on papers published in Web of Science, Scopus, Pubmed, and covered recommendations of public health protection and regulatory agencies around the world. The transmission of the severe acute respiratory syndrome (SARS-CoV-2) by food was not confirmed until the present time. In any case, the protocols already established for food safety were reinforced, emphasizing the proper hygiene of hands after shopping, handling food packages, or before manipulating or eating food, adequate social distance, the use of individual protection equipment, the health of employees, and the proper preparation of food. It is hoped, in the post-pandemic scenario, to reach a better understanding of the particularities that led to greater care with food hygiene. Moreover, it is expected that the food system will creatively adapt the way meals are served.

**Keywords:** Food quality control; SARS-CoV-2 pandemic; foodborne viruses; novel coronavirus; public health; safe food handling.

Souza TSP, Miyahira RF, Matheus JRV, Nogueira TBB, Santos-Maragoni C, Barros FFC, Antunes AEC, Fai AEC. Food services in times of uncertainty: Remodeling operations, changing trends, and looking into perspectives after the COVID-19 pandemic. Trends in Food Science & Technology 2022; 120(2022): 301-307. <https://doi.org/10.1016/j.tifs.2022.01.005>

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<b>Food services in times of uncertainty: Remodeling operations, changing trends, and looking into perspectives after the COVID-19 pandemic</b>	
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<b>ARTICLE INFO</b>	<b>ABSTRACT</b>
<b>Keywords:</b> Coronavirus outbreak Consumer food service operators Restaurants The “new normal” Eating behaviors Delivery	<b>Background:</b> Social distancing and the economic downturn imposed by COVID-19 have significantly affected the food service segment. Therefore, operation recovery and adapting to a new reality must be achieved as quickly and efficiently as possible. Studies on this topic, which have been conceptualized in various parts of the world, have brought new ideas to light to mitigate the negative effects of COVID-19 on food service. <b>Scope and approach:</b> This study aimed to discuss the impact of COVID-19 on food service operations, changes in pre-existing trends, and post-pandemic perspectives. <b>Key findings and conclusions:</b> COVID-19 has changed all business segments. When dining rooms were forced to close, many food services had to resort to innovation to survive, and many added deliveries and/or adopted the dark kitchen models in one of their many forms. It is expected that the demand for delivery, dark kitchens, and the adoption of technological solutions, for example, contactless payment, will remain in the post-pandemic scenario. Food quality control measures have become more strictly enforced, not only to prevent SARS-CoV-2 contamination but also to increase credibility with the customer. These long-established food safety practices have returned to the spotlight, been revised, and should be maintained for well into the post-pandemic period. Restaurants are operating again and restrictions on opening hours and capacity have been relaxed or eliminated. Continued studies on this topic are important for supporting creative and scientifically based solutions for socio-economic recovery.

Souza TSP, Matheus JRV, Barone AS, Ferreira DCM, Pelissari FM, Fai AEC. Development of biodegradable food packaging in the context of COVID-19: sustainability more urgent than ever. Sustainability Management Forum 2023; 30(Suppl 1): 1-11.

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SCIENCE-POLICY PERSPECTIVES 

## 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. This thesis paper discusses arguments from the literature regarding the current development of biodegradable food packaging in the context of COVID-19. It takes an international perspective concerning the impact of the pandemic and correlates it with the changes in current and post-pandemic perspectives. 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 focusing on recyclability. Besides that, there is a necessity to change the type of economy adopted nowadays—that is, the linear economy, to the circular economy.

**Keywords** Biodegradable food packaging · Circular economy · COVID-19 impacts · One single used plastic

Santos FH, Ferreira DCM, [Matheus JRV](#), Fai AEC, Pelissari FM (2024). Antioxidant Activity Assays for Food Packaging Materials. In: Otoni, C. (eds) Food Packaging Materials. Methods and Protocols in Food Science. Humana, New York, NY. [https://doi.org/10.1007/978-1-0716-3613-8\\_17](https://doi.org/10.1007/978-1-0716-3613-8_17)

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# Antioxidant Activity Assays for Food Packaging Materials

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 241 Accesses

### Abstract

Antioxidant packaging is an emerging technology that limits deteriorative reactions in oxidation-sensitive food products. The direct interaction of the antioxidant material with the packaged product may inhibit oxidation reactions by scavenging free radicals, consequently improving the food stability and extending its shelf-life. Although these packages represent a promising alternative for preserving food, until now, standardized procedures to accurately quantify their efficacy have been lacking. The methodologies employed to assess the antioxidant activity of food packaging are the same as those already used for natural extracts. These methods measure the ability of the analyzed material to scavenge free radicals. Herein, we describe in detail the principal methodologies that have been used to evaluate the antioxidant activity of food packaging materials.