Ciência Rural, Santa Maria, v.53:2, e20210679, 2023
http://doi.org/10.1590/0103-8478cr20210679
ISSNe 1678-4596
FOOD TECHNOLOGY

Gelatin-based polymeric films for applications in food packaging: an overview of advances, challenges, and perspectives

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ABSTRACT: *This research carried out a bibliometric analysis and literature review on the production of gelatin-based films for application as food packaging, addressing the main advances and limitations. The search for articles was performed in the Scopus database, and bibliometric data were obtained using the Bibliometrix tool (RStudio software). It was observed that a wide variety of compounds can be incorporated into gelatin films to overcome the limitations related to their high solubility and low mechanical properties, as well as to obtain active or smart functions. Among the most reported compounds were essential oils, pigments extracted from vegetables, and other antimicrobial agents. The most reported foods as an application matrix were meat (fish, chicken, and shrimp), milk, cheese, and minimally processed fruits. Even with promising trends, the biggest challenge for large-scale applications is to obtain easily degradable biopolymers with structural and functional stability similar to synthetic polymers. Thus, a greater focus on this theme in research may favor significant advances in the use of these packages and positively impact several of the Sustainable Development Goals, as recommended by the United Nations.*
Key words: *biopolymer, biodegradable packaging, sustainability, food conservation.*

Filmes poliméricos à base de gelatina para aplicações em embalagens de alimentos: uma visão geral sobre avanços, desafios e perspectivas

RESUMO: *Este estudo objetivou realizar uma análise bibliométrica e revisão de literatura acerca da produção de filmes à base de gelatina para aplicação como embalagens alimentícias, abordando os principais avanços e limitações. A busca de artigos foi realizada na base de dados da Scopus e os dados bibliométricos obtidos pela ferramenta Bibliometrix (RStudio software). Verificou-se uma grande variedade de compostos que podem ser incorporados nos filmes de gelatina, a fim de superar suas limitações relacionadas à alta solubilidade e baixas propriedades mecânicas, bem como para obtenção de funções ativas ou inteligentes. Dentre os compostos mais reportados, têm-se: óleos essenciais, pigmentos extraídos de vegetais e outros agentes antimicrobianos. Os alimentos mais reportados como matriz de aplicação foram: carnes (peixe, frango e camarão), leite, queijo e frutas, minimamente processadas. Mesmo com tendências promissoras, o maior desafio no âmbito das aplicações reais em larga escala é a obtenção de biopolímeros facilmente degradáveis, com estabilidade estrutural e funcional similar aos polímeros sintéticos. Dessa forma, o maior enfoque dessa temática em pesquisas poderá favorecer avanços significativos para o uso dessas embalagens, impactando positivamente diversos dos Objetivos para o Desenvolvimento Sustentável, preconizados pela Organização das Nações Unidas.*
Palavras-chave: *biopolímero, embalagens biodegradáveis, sustentabilidade, conservação.*

INTRODUCTION

Among the Sustainable Development Goals (SDGs) listed by the United Nations (UN), Zero Hunger (SDG 2) stands out for the food area, which advocates the eradication of hunger. To feed 10 billion people by 2050, it is necessary to strike a balance between sustainability, food security, food safety, and reduction in the wastage of food produced (VÅGSHOLM et al., 2020).

One way to reduce the wastage of food and strengthen the sustainability of the food cycle is to extend the shelf life of food products (BIANCHI et al., 2021). Among the available technologies, packaging is used to separate food from the environment, reduce exposure to spoilage factors, and prevent the loss of desirable compounds, thus prolonging the shelf life of food (OTONI et al., 2017).

However, traditional plastic packaging threatens the safety of the environment and human

Received 09.16.21 Approved 02.25.22 Returned by the author 04.19.22
CR-2021-0679.R1
Editors: Rudi Weiblen Jesús Lozano Sánchez

beings, boosting studies for the production of new composites based on biodegradable materials for applications in food industry (YAN et al., 2021). Biodegradable packaging is economical, safe, non-toxic, sensitive, and use natural pigments that can act as a quality indicator. In addition, these packaging films can be an optimized, marketed, and used as active and intelligent packaging materials, which helps in assessing the visual quality of food products (BHARGAVA et al., 2020).

One of the main biopolymers used for the preparation of biodegradable films is gelatin, a high molecular weight polypeptide, which is naturally present in different animal sources, including pig, cattle, and fish skin (PAHOFF et al., 2019). Gelatin is one of the most interesting biomaterials for the development of biopolymers, mainly due to its good film-forming properties and abundance in nature. However, its hydrophobic characteristics result in a rapid degradation process as the hydrogen bonds easily rupture in humid environments and temperatures above 30–35 °C, destroying the physical network of gelatin (ROSSETO et al., 2021; RIGUETO et al., 2021). Nevertheless, the addition of other compounds to the gelatin network makes it possible to obtain materials with improved characteristics that overcome the limitations of gelatin (RIGUETO et al., 2021).

In this context, this study conducted a bibliometric analysis and literature review on the production of gelatin-based films for application in food packaging.

METHODOLOGY

The survey of articles was carried out by searching for specific terms in the Scopus database. The terms (“GELATIN” AND “FILM” AND “PACKAGING”) were searched in the titles, abstracts, and/or keywords of the articles, yielding a total of 572 documents.

The results obtained in the preliminary search in the Scopus database were further narrowed down using the “Bibliometrix” tool (RStudio® software, version 1.4.1106), in which the search period was limited from 2016 to 2021 and only experimental articles were included (reviews, conference papers, and book chapters were excluded from the analysis), thus yielding a total of 392 articles.

These articles were analyzed for their content and most relevant results, and 31 articles (15 regarding active films and 16 regarding smart/intelligent films) were chosen to be covered in each topic. Thus, it was possible to systematize the state of the research and produce discussions and correlations pertaining to the theme of this review.

Figure 1 presents a flowchart that summarizes the methodology of this study.

DISCUSSION

Bibliometric analysis

In the bibliometric analysis, it was noted that the interest in the production and applications of gelatin-based films is growing, with an annual scientific production growth rate of 23.94%. There has been a marked growth in publications on this topic in 2021, with the highest peak of publications observed during this year (Figure 2a). The country that has published the most articles focused on the production of gelatin-based films as packaging food is China, followed by Iran, Brazil, Malaysia, and Thailand (Figure 2b).

From the WordCloud generated with the 50 most cited words in the keywords of the articles (Figure 2c), it is evident that the main applications of gelatin-based films as food packaging (see words circled in red) are mainly in the form of “edible films,” generally related to fruit and vegetable coating packaging, “food packaging,” “active packaging,” “smart packaging,” and “biodegradable films.” In addition, the words circled in yellow suggested some sources of gelatin extraction, such as “chicken skin gelatin” and “fish gelatin. The words circled in black, viz. “essential oil,” “sodium alginate,” and “curcumin,” indicated that these compounds have been added to the composition of gelatin-based films for technological purposes.

The following subtopics have been listed considering the main applications of gelatin-based films in food packaging, addressing the main advances and challenges reported in the selected studies.

Literature review

Active packaging

Active packaging is a system in which the product, packaging, and the environment positively interact to extend shelf life, in addition to maintaining or improving the condition of packaged food, through the incorporation of components that would release or absorb substances in the packaged food or in the environment around the food (BIJI et al., 2015).

Antioxidant agents can be used to inhibit the oxidation of lipids or proteins, while antimicrobials can be used to inhibit the growth of spoilage or pathogenic microorganisms. In addition,

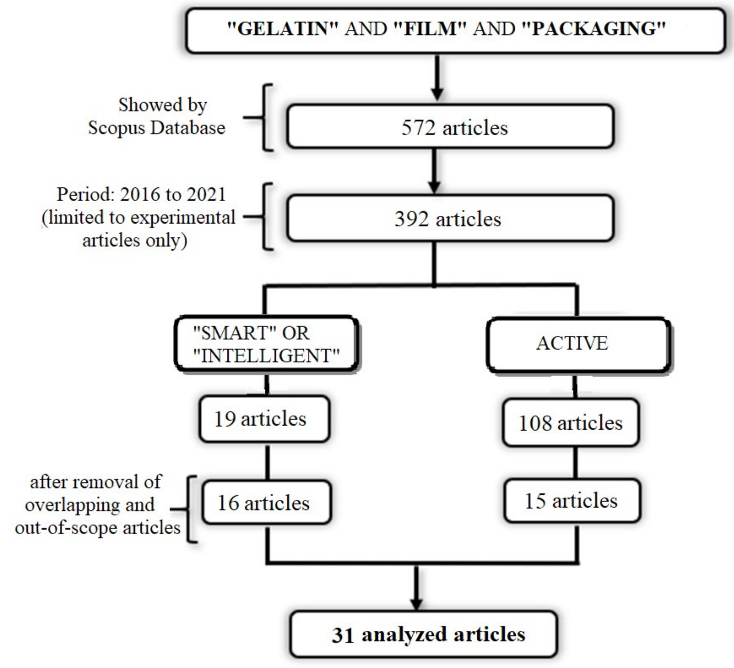

Figure 1 - Conditions established for the selection and analysis of articles included in this review.

substances that selectively absorb scattering ultraviolet or visible radiations can be incorporated to protect the photolabile food components (TAVASSOLI et al., 2021).

Figure 3 shows the main active agents that can be incorporated into packaging and/or hydrogels in the food industry.

Notably, a wide variety of active agents can be incorporated into the gelatin films. The choice must be based on the application of the films, considering that some additives shown in figure 3 can change the color and influence the sensory characteristics of the product. For example, in a study carried out by SETTIER-RAMÍREZ et al. (2021), films were made from gelatin and polyvinyl alcohol with the incorporation of *Lactococcus lactis* subsp. *lactis* for creamy mushroom soup and sliced ham. Regarding the sensory aspect, there were no changes in the cooked ham after 16 days, while consumers noticed acidification of the mushroom soup caused by a decrease in the pH.

Furthermore, it is necessary to evaluate the compatibility between the additive and the polymer matrix. In this context, gelatin stands out for usage in films because of its non-toxicity, lack of accentuation of flavor, and compatibility with different materials (SHAKILA et al., 2016).

Table 1 presents recent studies aiming the production of gelatin-based films with the incorporation of active agents applied in various foods.

Essential oils promote physicochemical improvements in the films and application matrix by reducing their interaction with water, which is reflected in lower water solubility and permeability to water vapor, in addition to antimicrobial resistance (FATHI-ACHACHLOUEIET et al., 2021). A study carried out by SHALILA et al. (2015) compared gelatin films (control) with gelatin films incorporating essential oils of chitosan, clove, and pepper, for their application in the packaging of fish fillets. After 6 days, the authors observed that the control film was completely decomposed with bacterial counts (*Staphylococcus aureus*, *Listera monocytogenes*, and *Aeromonas hydrophila*) at 10^7 CFU/g, while films with essential oils maintained a bacterial count of 10^5 CFU/g.

Another interesting factor is the possibility of using agroindustrial wastes as active ingredients,

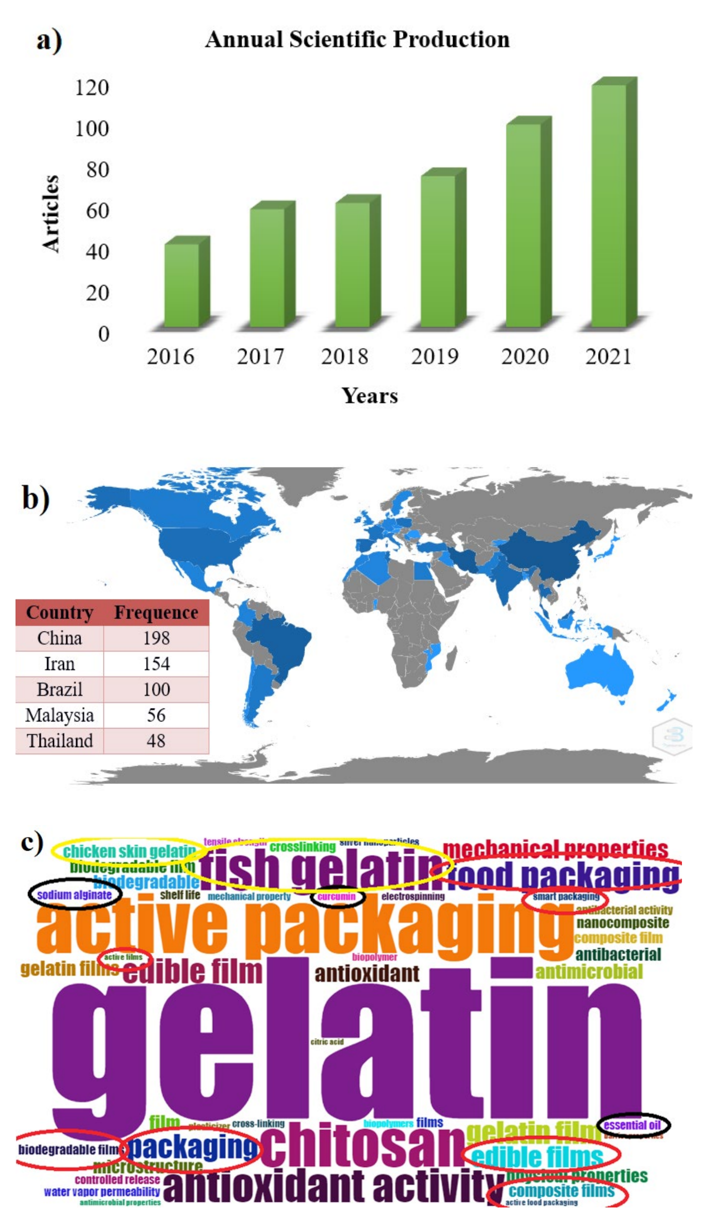

Figure 2 - a) Annual global scientific production of articles related to gelatin-based films; b) Scientific production of articles by country; c) WordCloud with the 50 most cited words in the keywords of the articles presented by the Scopus database.

Source: Bibliometrix – RStudio software (2021).

as exemplified in a study by SUSMITHA et al. (2021), who incorporated extracts from mango peels and pineapple bagasse to prepare starch and gelatin films. The authors reported that the addition of these components improved the thickness, moisture content, expansion index, and optical properties of the film, in addition to showing good material compatibility, as evidenced by FTIR analysis. HANANI et al. (2019) used pomegranate peels to prepare gelatin films and found it promising for the application as

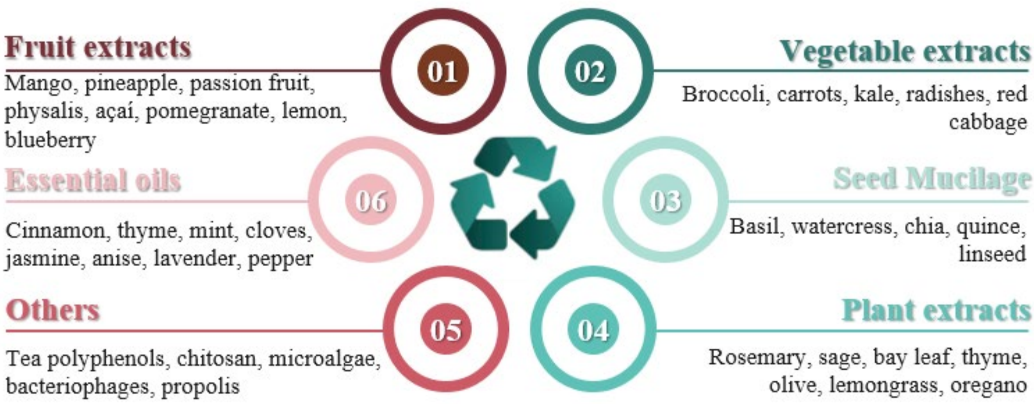

Figure 3 - Main active agents incorporated in food packaging and/or hydrogels.

active packaging because of its antimicrobial and antioxidant properties.

In addition, the use of Aloe vera (AV) extract has been widely reported for its applications in food packaging, owing to the high content of unique polysaccharides (glucomannan and acemannan), phenolic compounds (flavonoids and anthraquinones), organic acids, and vitamins (B_1 and B_2 among others); it yields homogeneous films with good transparency (PARVEN et al., 2020; SÁNCHEZ et al., 2020) and reduced solubility without causing a significant change in the thickness, color, and surface microstructure, in addition to improving the plasticity of the films (CHIN et al., 2017).

SANCHES et al. (2020), for example, added AV gel to fish gelatin-based edible films. The authors reported that the films had antimicrobial properties against *Staphylococcus aureus* and that AV at a concentration of 4% (w/w) could increase the postharvest quality of low-water content packaged foods, such as nuts, sprouts, and avocados, thus reducing the use of synthetic additives and contributing to the concept of circular economy by reducing food waste.

Smart/Intelligent packaging

Smart or intelligent packaging is defined as materials that monitor the state of the packaged food or surrounding external environment, and provide the consumer with information about the conditions of the food or its environment (temperature and pH); this communication is based on the ability to detect, sense, and record changes in the products' environment. Unlike active components, smart components are not intended to release their constituents in to the packaged food (BIJI et al., 2015).

A classic example of smart packaging is monitoring the pH of foods by visual colorimetric changes using natural pigments, such as anthocyanins, which are included in the matrix of the base component of smart packaging (ALPASLAN et al., 2020).

The use of gelatin as a base for manufacturing smart packaging is promising due to the possibility of obtaining composite materials; therefore, indicators can be added to the packaging material to improve mechanical strength and antioxidant and antimicrobial properties, among other benefits (MUSSO et al., 2016).

In this context, smart films are commonly used in food packaging because of their advantages, which include portable size, low cost, extended storage and shelf life of food, and real-time monitoring of food spoilage (CHAYAVANICH et al., 2020).

Table 2 presents recent studies that aimed the production of gelatin-based films for application as smart packaging in various foods.

As shown in table 2, ALPASLAN et al. (2020) and (2021), applied composite hydrogels based on gelatin with an aqueous extract of pomegranate peels and red apples, respectively. With the addition of aqueous apple extract, an improvement in the thermal stability and a reduction in the elasticity of the films were observed. Furthermore, when tested against *Escherichia coli, Bacillus subtilis*, and *Staphylococcus aureus*, the antimicrobial activity of the films was verified.

Table 1 - Production and application of films as active food packaging.

Active packaging	Performed or suggested applications	Reference
Gelatin films with anise essential oil incorporation	Chicken fillet	FATHI-ACHACHLOUEI et al. (2021)
Gelatin and polyvinyl alcohol films incorporating *Lactococcus lactis* subsp. *lactis*	Creamy mushroom soup and sliced cooked ham	SETTIER-RAMÍREZ et al. (2021)
Gelatin films with bacteriophage incorporation	Cheese	WENG et al. (2021)
Edible films of starch and gelatin with the addition of mango and pineapple extract	No application in specific foods	SUSMITHA et al. (2021)
Active packaging of fish gelatin with incorporation of protocatechuic acid	Chilled meat bovine	ZHONG et al. (2021)
Gelatin and agar-based film integrated with clove essential oil stabilized with nanocellulose	No application in specific foods	ROY & RHIM (2021)
Gelatin and chitosan-based films with the addition of soy straw nanocellulose and Pitanga extract (*Eugenia uniflora* L.)	No application in specific foods	TESSARO et al. (2021)
Gelatin film of porcine skin and zein with the addition of ethanolic propolis extract	Raspberry	MORENO et al. (2020)
Fish gelatin films with orange peel pectin incorporation	Cheese	JRIDI et al. (2020)
Fish gelatin films with pomegranate skin	No application in specific foods	HANANI et al. (2019)
Gelatin and cassava starch films with curcumin incorporation	Sausages	TOSATI et al. (2018)
Influence of starch oxidation on the functionality of starch-gelatin based active films	No application in specific foods	MORENO et al. (2017)
Chitosan and gelatin films containing *Ziziphora clinopodioides* essential oil, pomegranate peel extract, and cellulose nanoparticles	Peeled shrimp	MOHEBI & SHAHBAZI (2017)
Mixed films of pigskin gelatin and sodium caseinate with extracts of boldo, guarana, cinnamon, and rosemary	No application in specific foods	BONILLA & SOBRAL (2017)
Fish gelatin films incorporating chitosan, clove, and pepper essential oils	Fish fillet	SHAKILA et al. (2015)

The addition of citric acid and dimethyl acrylamide improved the mechanical properties, while when using the pomegranate extract, anthocyanins and natural antimicrobial and antioxidant properties were obtained (ALPASLAN et al., 2021). Both studies applied the obtained hydrogels in dairy products (milk and cheese). Since, when whole milk and cheese reach temperatures above 4 to 7 °C, certain changes can occur, leading to a change in the pH of the medium. The authors reported that hydrogels were effective in accurately detecting the pH changes in the tested dairy products, thus proving to be promising candidates for use as biodegradable materials in food packaging.

Gelatin films with red radish extract, produced by CHAYAVANICH et al. (2020), were applied to chicken and shrimp samples. The authors evaluated the color stability of the detection films, and the results indicated that the films had good stability. However, the degradation was higher during the storage of the samples containing the smart packaging at room temperature than that at refrigeration and freezing temperatures, which allowed for storage for more than two weeks; for both samples, the films used were pH-sensitive for real-time observation of meat deterioration with good accuracy.

The solvent used in the extraction of natural pigments, such as anthocyanins, can also directly interfere with the properties of the films, as reported by MUSSO et al. (2019) The alcoholic extracts of red cabbage were more effective than the aqueous extracts because of the greater concentration and variety of anthocyanins, which exerted a plasticizing effect on the gelatin protein matrix, causing an increase in the elongation and solubility of the films. Conversely, the authors emphasized that aqueous extracts seemed to favor protein crosslinking, as they improved the mechanical behavior of the films.

In addition to pigments extracted from natural sources, studies have reported the use of commercial pH indicator compounds; MUSSO et al. (2016) added three acid-base indicators to gelatin films: methyl orange (pH=2), neutral red (pH=6), and bromocresol green (pH=11). The authors observed

Table 2 - Production and application of films as smart/intelligent food packaging.

Smart packaging	Performed or suggested applications	Reference
Intelligent gelatin packaging films with lavender essential oil Pickering emulsions and Alizarin	Shrimp	WANG et al. (2022)
Hydrogel made from poly (gelatin-co-dimethyl acrylamide) with added citric acid and red apple peel extract	Pasteurized milk and cheese	ALPASLAN et al. (2021)
Bio-amine responsive films based on starch/glycerol/gelatin enriched with colored potato (Black King Kong) anthocyanin	Pork freshness	NIU et al. (2021)
Gelatin films with addition of curcumin, betanin, and anthocyanin	Fatty food	ETXABIDE et al. (2021)
Gelatin films with addition of anthocyanin from butterfly pea extract	No application in specific foods	RAWDKUEN et al. (2020)
Hydrogel containing poly (dimethyl acrylamide), gelatin and pomegranate extract (*Punica granatum*)	Whole milk and cheese	ALPASLAN et al. (2020)
Gelatin-carrageenan-based film integrated with shikonin and propolis	Milk	ROY & RHIM (2020)
Gelatin/oxidized chitin nanocrystals nanocomposite films containing black rice bran anthocyanins	Fish	GE et al. (2020)
Alginate/starch/gelatin composite films with addition of red cabbage extract	Sheep meat	ALVES et al. (2021)
Composite films from starch, gelatin, glycerol, acetic acid, and curcumin	No application in specific foods	NGUYET & NGUYEN (2020)
Permeable edible film based on κ-carrageenan, gelatin, and curcumin	Grass carp fillets	HE et al. (2020)
Gelatin and starch film containing red radish extract (*Raphanus sativus* L.)	Chicken and Shrimp Meat	CHAYAVANICH et al. (2020)
Gelatin film with red cabbage (*Brassica oleracea* L.) extract	No application in specific foods	MUSSO et al. (2019)
Gelatin film with addition of aqueous hibiscus extract	No application in specific foods	PERALTA et al. (2019)
Films based on starch/gelatin nanoparticles	Peeled and sliced apples	TAO et al. (2018)
Gelatin film containing acid-base indicators	No application in specific foods	MUSSO et al. (2016)

rapid visual response of films due to the changes in the pH of liquids and gases; however, for semisolids, the response was slower, possibly due to limited diffusion processes. Regarding the composition of the films, the addition of methyl orange and neutral red resulted in physical and/or chemical crosslinking, increasing the tensile strength and reducing the water solubility of the films, without affecting their permeability to water vapor and their ability to change their color in relation to the pH of the surrounding medium.

Given that milk is a nutritious food that is widely consumed by the human population, it would be of extreme importance for the consumers if, during storage in homes or supermarkets, the freshness of the milk could be monitored by visual change using a pH-based color sensor. In this context, ROY & RHIM (2020) evaluated gelatin and carrageenan films with added shikonin and propolis to monitor milk freshness. The authors concluded that the mixture of propolis and shikonin improved the UV blocking property of the films, without significantly decreasing the transparency, and imparted antimicrobial and antioxidant activities to the films; however, water resistance and expansion rate increased. The color indicator films showed excellent pH response, with color change observed over a wide pH range from 2 to 12, thus making it a promising candidate, which can be increasingly used in packaging for monitoring the freshness of the packaged food.

TAO et al. (2018) pointed out that the application of films on peeled apples caused an additional resistance to mass transfer, and the coatings decreased the diffusion rate of water molecules, which may be related to the edible coating acting as a physical barrier, causing a reduction in the respiration rate of fresh vegetable products.

In addition, we observed that in most studies that produced gelatin-based films, essential oils were used either alone or in blends to provide active properties to the films. However, a recent study by WANG et al. (2022) showed the possibility of producing intelligent active gelatin films to maintain and monitor shrimp freshness simultaneously. It used lavender essential oil Pickering emulsions and

Alizarin (as pH indicators and biogenic amines) integrated into the three-dimensional (3D) network of the gelatin matrix via intermolecular hydrogen bonding and dehydration condensation, showing sensitivity to color change and rapid response.

An overview of challenges and social aspects

Even with promising trends in food applications, gelatin-based edible packaging materials have limitations that need to be overcome in order for real and large-scale applications. For example, the mechanical properties (elongation and tensile strength) are associated with the performance of these materials in keeping themselves intact during the stress caused by processing, handling, and/or storage; hence, the objective is to obtain resistant and flexible materials (JIANG et al., 2019; SHI et al., 2017).

Water-resistance is a necessary property for films intended for use as food packaging. As gelatin has water-sensitive properties and the films produced from it dissolve easily in aqueous media, its applications in many fields are limited. Therefore, promoting the hydrophobization of gelatin films is essential to realize real applications of these packages (ALPASLAN et al. 2020; TAO et al., 2018).

Furthermore, the water vapor permeability of edible films and their angle of contact significantly affect the physical and barrier properties; therefore, it is necessary to obtain films with standardized water content and contact angles (TAO et al., 2018).

In general context, the main challenge for widespread applications of gelatin-based films as food packaging is to obtain easily degradable biopolymers; however, their structural and functional stability needs to be similar to that of synthetic polymers.

With regard to social aspects, taking into account the SDGs of the UN, the use of gelatin-based films for applications, such as food packaging, can positively impact several of these goals, as illustrated in figure 4.

The positive impact on SDG 2 (Zero Hunger) is due to the food preservation technology employed, ensuring more food is available to the population, while on SDG 3 (Good health and Well-being) is because the packages have antimicrobial and antioxidant properties, which, in addition to monitoring the degree of food deterioration, can inhibit microbial growth, thus ensuring safe food for the consumers. The positive impact of SDG 9 (Industry, Innovation, and Infrastructure) is due to the opportunities for innovation by industries, guided by the appeal of new food trends demanded by the consumers, and that of SDG 12 (Responsible consumption and production) is due to the consumption of biodegradable products from sustainable sources. The positive impacts on SDG 14 (Life below water) and SDG 15 (Life on land) are due to the accelerated degradation of packaging, reducing the environmental impact.

CONCLUSION

In this research, a bibliometric analysis and literature review of articles pertaining to gelatin-based films used as active and smart packages for applications in the food industry are presented. The main foods used in the applications and the most

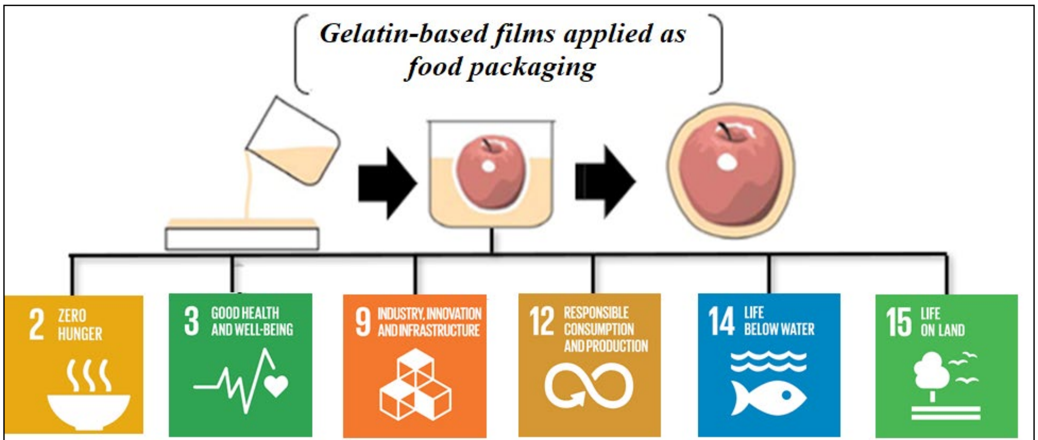

Figure 4 - Main SDGs that can be positively impacted by the use of gelatin-based films as food packaging materials.

promising results of the analyzed studies are reported. In addition, the limitations that need to be overcome for real and large-scale applications, in addition to the positive impacts of these applications on society, have been discussed in a generic way.

The addition of compounds with active or intelligent properties to the composition of gelatin films favors the improvement of certain properties; for example, essential oils impart hydrophobic characteristics and tend to reduce the solubility or water vapor permeability, in addition to conferring antimicrobial capacity and increasing the shelf life of the product. However, it is necessary that studies include the sensory analysis of edible films applied to the evaluated foods, since certain compounds can impart undesirable odors or flavors to the product, resulting in a rejection by the consumers.

We believed that a greater focus on research to obtain gelatin-based films with improved properties may favor significant advances in the use of these packaging, directly contributing to scientific and technological development, preservation of the environment, and reduction in food losses.

ACKNOWLEDGEMENTS

The authors would like to acknowledge the Coordenação de Aperfeiçoamento de Pessoal de Nível Superior (CAPES) – Finance Code 001.

DECLARATION OF CONFLICT OF INTERESTS

The authors declare no conflict of interest. The founding sponsors had no role in the design of the study; in the collection, analyses, or interpretation of data; in the writing of the manuscript, and in the decision to publish the results.

AUTHORS' CONTRIBUTIONS

The authors contributed equally to the manuscript.

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