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Nanotechnology innovation in bioactive packaging for perishable foods - a review

Inovação nanotecnológica em embalagens bioativas para alimentos perecíveis – uma revisão

Lilian Raquel Moretto Ferreira[1], Ligia Chitolina, Indianara Cristina Dias, Creciana Maria Endres, Marcia Adriana Tomaz Duarte.

[1]Centro Universitário SENAI/SC

[*]Correspondente: lilian.ferreira@edu.sc.senai.br

Abstract

This paper provides a review of the use of nanotechnology in the primary packaging of chilled meat foods through the electrowinning of biodegradable polymers by incorporating essential oil. The main raw material for packaging production comes from non-renewable sources such as oil. However, synthetic polymers from this raw material generate a large volume of solid waste, making it essential to search for new alternatives to supply the packaging market and decrease the environmental and ecological impact. The latest data from the Food and Agriculture Organization of the United Nations corroborate this problem by pointing out that 127 million tons of food are discarded every year in Latin America, and many foods are wasted because they are not appropriately packaged and thus without the guarantee of safe storage and transportation conditions. Food losses along the production and supply chains must be halved to contribute to the Sustainable Development Goals by 2030. Thus, developing the national packaging industry for perishable foods is under discussion and motivates entrepreneurship, as it uses a significant amount of packaging. Moreover, it is increasingly expected to find sustainable alternatives in production lines to reduce environmental impacts and provide gains for packaged perishable products. Nonetheless, new studies must present the technologies that can be used, as well as the results and limitations of their application. These data can contribute to the production and scalability of these packages in industrial environments.

Keywords: bioactive packaging; food; eugenol; electrowinning; essential oils; nanoparticles.

Resumo

Este trabalho traz uma revisão sobre o emprego da nanotecnologia nas embalagens primárias dos alimentos cárneos refrigerados por meio da eletrofiação de polímeros biodegradáveis com incorporação de óleo essencial. A principal matéria prima para a produção de embalagens, vem de fontes não renováveis, como o petróleo, porém, os polímeros sintéticos provenientes desta matéria prima geram um grande volume de resíduos sólidos, assim, torna-se fundamental a busca por novas alternativas para suprir o mercado de embalagens e desempenhar um papel de menor impacto ambiental e ecológico. Os últimos dados da Organização das Nações Unidas para a Alimentação e a Agricultura corroboram com está problemática apontando que 127 milhões de toneladas de alimentos são jogadas fora por ano na América Latina e muitos alimentos são desperdiçados por não estarem embalados adequadamente e assim sem a garantia de condições para um armazenamento e transporte seguro. Para contribuir com os Objetivos do Desenvolvimento Sustentável (ODS) até 2030, deve-se reduzir pela metade as perdas de alimentos ao longo das cadeias de produção e abastecimento. Assim, o desenvolvimento da indústria nacional de embalagens para alimentos perecíveis está em discussão e motiva o empreendedorismo, pois essa utiliza uma quantidade significativa de embalagens. Além de que cada vez mais espera-se encontrar alternativas sustentáveis nas linhas produtivas, a fim de reduzir os impactos ambientais e proporcionar ganhos para os produtos perecíveis que são embalados. Porém é necessário, que novos estudos apresentem as tecnologias que podem ser utilizadas, bem como os resultados e limitações da sua aplicação. Esses dados poderão contribuir para a produção e escalabilidade dessas embalagens em ambientes industriais.

Palavras-chave: embalagens bioativas; alimentos; eugenol; eletrofiação; óleos essenciais; nanopartículas.

1. INTRODUCTION

The demand for food has increased due to population growth and how modern society consumes, which also implies an increase in plastic waste from packaging (RUVIARO *et al*., 2020). A study commissioned by the the Plastic Chain Incentive Plan (PICPlast) reported that only 23.1% of post-consumption plastic waste in Brazil was recycled in 2020. Therefore, most of it is composed of non-biodegradable waste and remains on our planet for many years, compromising human health and the environment.

Most of this waste comes from traditional petroleum-based commercial food packaging materials, such as polyethylene (PE), polypropylene (PP), and polystyrene (PS). Researchers estimate that 31.9 million tons of plastic waste enter the environment each year, with 4.8-12.7 million tons going into the oceans in amounts that are enough to contaminate terrestrial ecosystems (KAWECKI *et al*., 2019). Still, the demand for plastics is expected to continue growing in the future to enable resource-efficient products needed by society.

Nonetheless, one of the trends is the development of recycled products and renewable raw materials that use renewable energy-driven processes to establish an efficient circular economy system. According to the UN Sustainable Development Goals and the Sustainable Plastics Strategy, a shift in the global plastics industry is necessary, replacing a manufacturing system based predominantly on fossil fuels with sustainable and affordable alternatives (LAVRIČ *et al.* 2021). Currently, most of the packaging used in the food industry is made of plastic, as it has the classic functions of containing, protecting, and selling packed products. The protective function involves preserving the quality of the product as much as possible by creating conditions that minimize chemical, biochemical, and microbiological changes that cause degradation. Traditional packaging requires improvements to extend the shelf life of food products and meet the growing consumer demand for safe, healthy products closer to natural ones and with fewer preservatives (SOARES *et al.,* 2009).

It is estimated that the annual production of plastic is 200 million tons worldwide, derived from petroleum, and not biodegradable, presenting a huge environmental problem (COSTA, 2011). In order to reduce the environmental impacts arising from the accumulation of plastic from synthetic polymers, research has sought to produce packaging made from polymers from renewable sources and that degrade over time through natural mechanisms.

The plastics used in the packaging represent an essential part of the processing of products, and another related issue is the waste of food in the consumption stage because of the packaging, which often does not fulfill its function of storing and protecting the product against extrinsic factors such as light, moisture, air, microbial agents, and others that may interfere and impair its quality. However, guaranteeing safe transportation, adequate packaging, and convenience for the consumer is paramount to the demanding market. In this way, more and more products that present benefits and conveniences for both the industry and the final consumers are the ones that receive more prominence (YAM, TAKHISTOV, & MILTZ, 2005).

In this context, bioactive packaging presents great growth. It highlights technologies that can reduce or even eliminate problems related to food deterioration, such as microorganism growth and chemical changes caused by oxidation, consequently increasing the shelf life of these products and presenting biodegradable and sustainable behavior (MIHINDUKULASURIYA & LIM, 2014; OTONI et al. 2016. The concept of bioactive packaging brings technologies in which the biodegradable packaging material interacts with food, aiming for benefits and actively combating deleterious agents (OTONI et al. 2016). Among the applications and purposes of bioactive packaging are those with antimicrobial activity, which inhibits or retard the growth of microorganisms (fungi, bacteria and viruses), contributing to preserving food's quality and shelf life. The antimicrobial properties of most primary packaging materials are based on the migration of active substances from the packaging to the food, then exerting its action (NERIN *et al.*, 2016).

A problem recently brought to the forefront by industry is the high resistance to antibiotics or antimicrobials that microorganisms acquire with their common use (LIU et al., 2015). With this comes the importance of the discovery and use of new antimicrobial agents of natural origin, including essential oils combined with metal nanoparticles, which is highlighted in the medical field and all areas, including food. The antimicrobial action of essential oils and zinc nanoparticles is proven in various studies regarding the inhibition of the following microorganisms: *Escherichia Coli, Salmonella enterica, Listeria monocytogenes, Listeria innocua, Penicillium toqueforti, Aspergillus flavus, Endomyces fibuliger*, and *Pseudomonas putida* (SU CHA et al., 2002; KECHICHIAN, 2007; SOARES et al., 2009; ZHANG, 2016). Antimicrobial packaging slows or even inactivates microbial growth, reducing the levels of synthetic preservatives applied to food. In this panorama, essential oils and nanoparticles as active substances represent an efficient and safe alternative for the composition of bioactive packaging because they are employed on a nanometer scale, enabling biodegradable packaging that increases the shelf life of foods. It also reduces the total environmental impact, especially for products with high losses, as is the case of perishables.

2. METHODOLOGY

To address nanotechnological innovation applied to a commercial product, searches were conducted in databases available on the internet in the last three years (i.e., from 2019 to 2022). This bibliometric review used distinct databases concerning the publication type, one being patents and the other with academic publications. The search terms and the respective databases are listed in Table 1. The results are represented by the number of publications found to make it possible to point out trends in the theme.

Table 1 - Amount of publications in the period 2019-2022

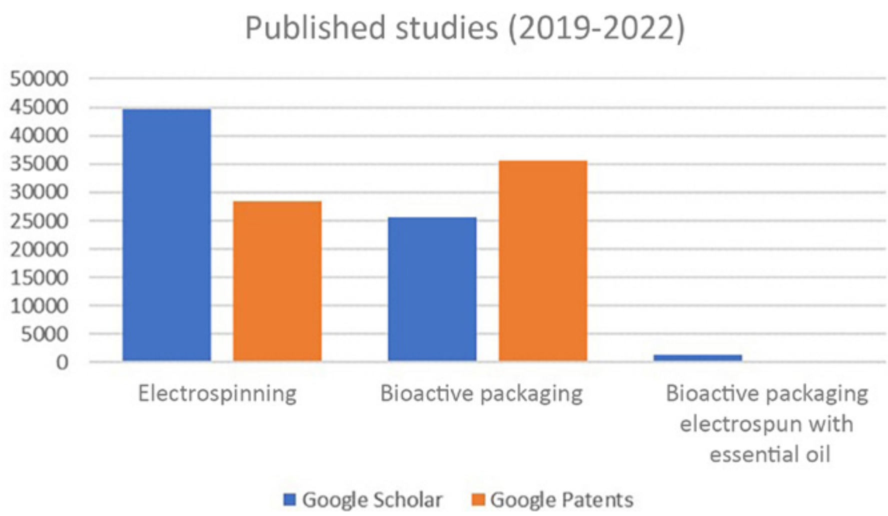
SEARCH TERM	GOOGLE SCHOLAR	GOOGLE PATENTS
Electrospinning	44,100	28,355
Bioactive Packaging	21,500	35,563
Bioactive Packaging	4,130	141
Electrowinning	686	31
Electrowinning of bioactive packaging	129	3
Electrowinning bioactive packaging essential oil eugenol	25	1
Bioactive packaging by electrospinnig	279	0
Electrospun Nanofibers Food Packaging	17,000	300
Electrospun Nanofibers Food Packaging essential oil eugenol	1,310	7

Source: The authors (2022)

From the Google Scholar database, which has scientific articles, master's or doctoral theses, abstracts, university journals, and books, we observed that when the search is conducted in English, we have 44,100 papers against 686 papers in the Portuguese language. Another relevant data is about the application of nanotechnology of electrowinning for bioactive packaging, where it is observed that of the 44,786 papers that deal with the technique of electrowinning, only 17,279 (38%) are intended for this technological application, and 1,310 (3%) papers present results for the use of essential oils, specifically the bioactive component eugenol, directly in the packaging material.

In the Google Patents database, we searched for granted patents, considering in this case that there was proof of invention of the technology from a new product or a new process. This search is essential when it comes to innovation because it protects the marketing rights of the interested party in the case of improvements in the use or manufacture of packaging. Thus, it can be seen that when the search for bioactive packaging is performed with the terms in English and Portuguese, the number of publications on Google Patents is 35,704. This number is higher than the number of publications on Google Scholar, which is 25,630, showing the preferential interest of the authors of this content in patent registration. The same behavior is not observed when the search term refers to electrowinning, in which 44,786 results are obtained at Google Scholar and 28,386 results at Google Patents. These results can be seen in Figure 1.

Figure 1 - A comparison chart of the quantitative results obtained for the search performed in the different databases



Source: The authors (2022)

By refining the search for the specific theme of electrowinning of bioactive packaging using the essential oil (eugenol compound) in the process of obtaining the packaging, a result is a significant number of works in Google Scholar, 1335, contrasting with a modest number of 8 in Google Patents. This demonstrates that the vast majority of works to be reviewed in this theme are in academic databases.

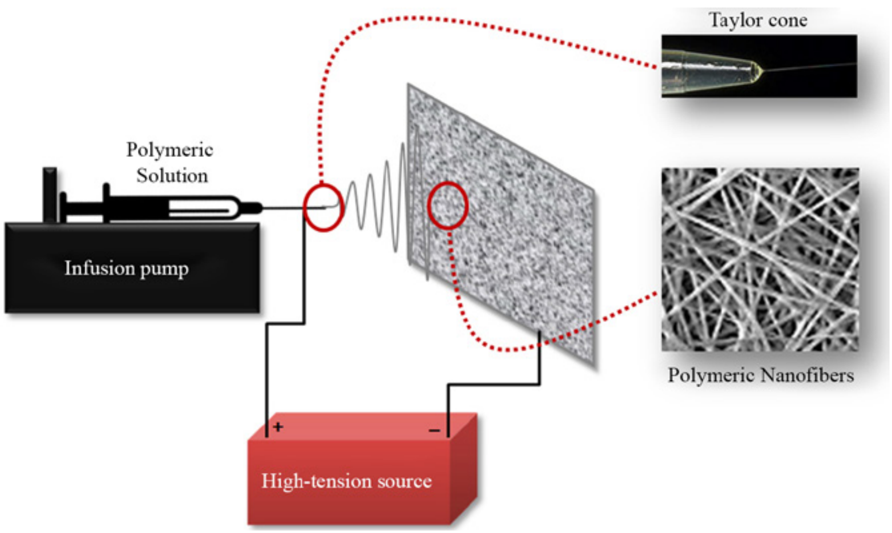
From this quantitative analysis in the databases, some papers were selected that made it possible to present evidence about the technological innovation that motivates entrepreneurship in bioactive packaging. These selected papers and others published in different periods make up the review that will be given below.

3. THE TECHNOLOGY OF ELECTROWINNING IN BIOACTIVE PACKAGING MANUFACTURING

Cooley and Morton first patented electrowinning in 1902 as a direct method to produce continuous micro and nanofibers. However, the elementary principles for obtaining nanofibers emerged through the junction of research conducted by John Zeleny in 1914, who evaluated the influence of electric force on the surface of liquids that had their surface changed. In 1964, Sir Geoffrey Taylor observed that viscous conductive fluids form thin jets by forming a cone called the Taylor cone when subjected to electric force and potential difference.

Electrowinning occurs when the load applied to the polymer solution is greater than the surface tension of the liquid; it deforms into a cone called a Taylor cone and is ejected in the form of a thin jet. The solvent from this jet evaporates and the membrane-shaped polymer from nanofibers with nanometer to micrometer diameter is deposited on a grounded collector (DOSHI; RENEKER, 1995). Figure 2 schematically illustrates the electrowinning system with its four basic components: syringe with the polymer solution, capillary tube with a metal injector, high voltage source, and the grounded collector plate where the polymeric nanofibers are formed. There are two sets of parameters that affect the morphology of nanofiber electrowinning: intrinsic parameters (such as solution viscosity, solvent evaporation rate, and polymer conductivity) and solution and processing parameters (which include voltage, feed rate, collector shape and texture, and collector distance).

Figure 2 - Schematic of the typical electrowinning process of packages/nanofibers



Source: Adapted from Ataei *et al*. (2020)

The nanotechnological innovations that permeate the electrospinning technique in bioactive packaging are related to the manufacturing process. This involves systematically adding antimicrobial agents directly into the polymer solution, followed by electrowinning. Although the incorporation of essential oils and nanoparticles into nanofibers has been performed previously in other industrial segments, employment in food packaging is relatively recent (JOUKI, 2014).

As a matrix used in electrowinning, natural polymers available on the market can be of plant, animal, or microbial origin; their properties are likely to be altered by different physical and chemical methods. This allows one to select desired properties such as water absorption capacity, degradation kinetics, and mechanical properties with appropriate specifications for certain applications (MATSUI, 2007). Polyvinyl Alcohol (PVA) is a biocompatible and non-toxic polymer that has been widely applied in the preparation of electrophilic antimicrobial materials (AYTAC, DOGAN, TEKINAY, & UYAR, 2014; WANG, YUE, & LEE, 2015). Polylactic acid (PLA), obtained from dextrose (sugar) extracted from renewable source materials, is one of the most promising biopolymers due to its properties with polyolefins with potential use in various fields such as food, drug delivery systems, and medical applications. Some authors question its barrier properties (RADUSIAN, 2016); however, this disadvantage can be overcome by employing nanotechnology, incorporating a small number of nanoactives into the polymer matrix and producing hybrid materials. Thus, nanostructured PLA has good physical-mechanical properties (RAMOS et al. 2016), is resistant to oil and water vapor, and has long transmittance. It has a good market price compared to other biodegradable polymers. Recently, the Food and Drug Administration (FDA) has approved PLA as a safe material to be in contact with food (Wen *et al.* 2016). Three-dimensional spherical nanoparticles (SiO_2, TiO_2, ZnO) showed great potential in improving the properties of PLA material. Pilić *et al.* (2015) reported that a meager amount of hydrophobic silicon oxide nanoparticles (0.2%) improve barrier properties up to 50% and show significant improvements in mechanical properties (tensile strength), which is explained by the type of nanoparticles, their good dispersion, and distribution in the PLA matrix. Wen *et al.* (2016) reported improvements in the thermal and mechanical properties of pure PLA by adding low content (up to 5%) of hydrophilic silicon oxide nanoparticles. In the work of Bittencourt *et al.* (2019), one can find a description of the manufacturing technique of polymeric nanofibers produced by electrowinning nanotechnology incorporated with essential oils in their polymeric structure.

3.1 Eugenol as an antimicrobial agent in bioactive packaging manufacturing

To add antimicrobial properties to PLA, a solution preparation of essential oils is performed before the electrowinning process. However, antimicrobials characterized as natural, efficient, and non-toxic agents are preferred due to health and ecological concerns (SUNG *et al*., 2013). Essential oils are natural substances with powerful antimicrobial activity against a wide variety of pathogens and are categorized as Generally Recognized as Safe by the FDA (BURT, 2004), indicating that they can be used in the food industry without additional approval because they are volatile organic and plant-based products obtained by a physical process. There is evidence that about 35% of plant essential oils have antimicrobial activity, and 65% have antifungal activity, acting in the conservation of the product (STIEVEN *et al*., 2009; LIMA *et al.,* 2006).

Antimicrobial agents can be incorporated directly into the polymeric matrix on labels and tags or be contained in sachets (OLIVEIRA & OLIVEIRA, 2004). Its addition in polymeric films can be done in two ways: incorporation and immobilization. In the first case, the antimicrobial agent is released into the food, while in the immobilization, the compound acts only at the surface level (HAN, 2005). The use of packaging containing antimicrobial agents has the advantage of diffusing these compounds to the food surface in a controlled manner. Thus, they are present in smaller quantities, meeting a current consumer demand, which is the search for preservative-free foods, and only where their presence is required (i.e., especially on the product's surface, where most deterioration occurs).

One of the promising antimicrobials of natural origin in the food industry can be found in eugenol (4-allyl-2-methoxyphenol), a natural phenol in many botanical extracts, such as essential oils of clove, cinnamon, and nutmeg. Eugenol is a colorless or slightly yellowish oil with a characteristic odor. (MURATORE *et al.*, 2019; FARMAKOLOJIK & ÖZELLIKLERI, 2017). For these reasons, it has sparked academic interest in research in applications in food packaging (AMORIM, 2019).

When evaluating the antioxidant and antimicrobial potential of cloves and their components, researchers have concluded that their use can prevent or reduce lipid oxidation, spoilage agents, and pathogens in food products, thus becoming an effective alternative natural preservative that extends the shelf life of products (GÜLÇIN et al., 2012; EL-MAATI et al., 2016). The presence of eugenol in nanofibers has been extensively investigated, showing antibacterial, antioxidant, and insecticidal activity. This has been reported in many papers confirming that the essential oil can effectively inhibit *E. coli* (Gram-negative bacteria) (REQUENA, VARGAS, & CHIRALT, 2019) and *S. aureus* (Gram-positive bacteria) (MURATORE *et al.*, 2019, CHENG et *al.*, 2019).

According to Geisse (2019) work, the essential oil of clove presents satisfactory results regarding its antimicrobial effectiveness, being proven for this purpose. Thus, eugenol becomes an attractive alternative to be inserted in active packaging for food with low cost, high yield, and effectiveness. When the antimicrobial agent is released from the packaging over time, the kinetics of microbial multiplication and antimicrobial activity on the surface of the perishable product is balanced until the growth of these products is reduced. In this way, it can be said that the antimicrobial activity of the packaging is extended, ensuring safety during food distribution.

3.2 The characterization of the antimicrobial activity of the packaging

The antimicrobial activity of essential oils is clear, although their mechanisms of action are not yet completely elucidated. There is consensus that most compounds are aromatic and phenolic and exert their antimicrobial effects directly on the cytoplasmic membrane, causing changes in the structure and functions of microorganisms (HOLLEY & PATEL, 2005). The characterization of the antimicrobial activity of packaging is performed in certified laboratories, most often following the ISO-BS 21702:2019 standard (Measurement of antiviral activity on plastics and other non-porous surfaces) and the qualitative disk diffusion method (WEN et al., 2016). Generally, studies have provided evidence of the efficiency of one or more gram-positive and gram-negative microorganisms such as *Staphylococcus Aureus, Escherichia Coli, Salmonella abony*, and *Listeria Monocytogenes*. Laboratory analyses are always performed using ATCC strains.

Figure 3 - Samples of bioactive packaging being prepared for the characterization of antimicrobial activity, being: (a) Addition of the microorganism in the packaging; (b) Culture medium

Source: The authors (2022)

The analysis results are expressed in mm of the zone of inhibition. To evaluate the nanofibers, they are placed in the lids of the Petri dishes, and the results are expressed in the percentage of reduction of bacteria growth.

4. CONCLUSIONS

Technological processes in the food industry that promote the reduction in the use of petroleum-based plastics with the benefit of safely extending the shelf life of perishable foods in packaging are becoming increasingly common. In this study, combining the electrowinning technique and the impregnation with essential oils for bioactive packaging production is a technological innovation capable of overcoming the limitations of conventional packaging production methods that compromise sustainability. Moreover, the new packages that are increasingly important in the market aim to achieve some main objectives, including extending the shelf life of food with quality and safety, reducing food waste, and reducing the addition of artificial preservatives or replacing them with natural substances with antimicrobial function.

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