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Review

Biobased Food Packaging Systems Functionalized with Essential Oil via Pickering Emulsion: Advantages, Challenges, and Current Applications

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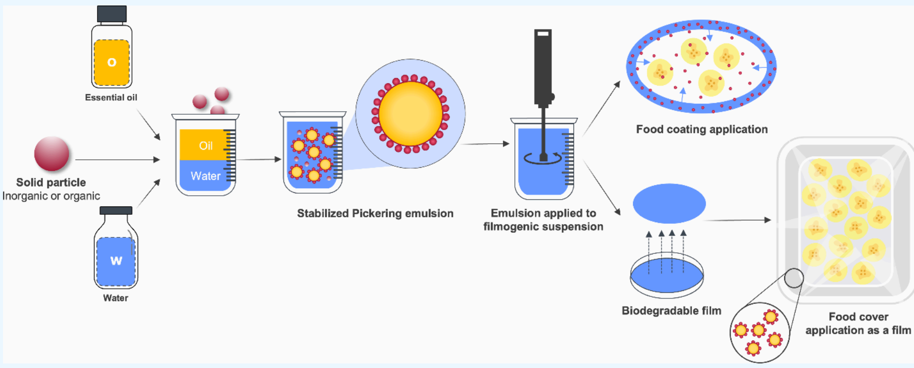
Cite This: https://doi.org/10.1021/acsomega.4c09320 Read Online



ABSTRACT: The development of innovative active food packaging is a promising strategy to mitigate food loss and waste while enhancing food safety, extending shelf life, and maintaining overall quality. In this review, Pickering emulsions with essential oils are critically evaluated as active additives for sustainable food packaging films, focusing on their antimicrobial and antioxidant properties, stabilization mechanisms, and physicochemical performances. A bibliometric approach was used to contextualize the current research landscape and new trends. Data were collected from the Web of Science and Scopus databases to find studies published between 2020 and 2024. The analysis of 51 articles shows that cinnamon, clove, and oregano are the most used essential oils, while cellulose and chitosan are the predominant polymer matrices. Pickering emulsions as stabilizers in food science represent a step forward in sustainable emulsion technology. The incorporation of essential oils into biobased films via Pickering emulsions can improve the mechanical and barrier properties, antimicrobial and antioxidant activities, and shelf life of foods. This approach offers a natural, environmentally friendly alternative to conventional materials and is in line with the 2030 Agenda goals for sustainability and responsible consumption. Recent advances show that composite particles combining polysaccharides and proteins have higher stability and functionality compared to single particles due to their optimized interactions at the interfaces. Future research should focus on developing scalable, cost-effective production methods and conducting comprehensive environmental testing and regulatory compliance, particularly for nanotechnology-based packaging. These efforts will be crucial to drive the development of safe and effective biobased active food packaging.

1. INTRODUCTION

The Food and Agriculture Organization (FAO) estimates that approximately one-third of the edible food produced globally for human consumption—equivalent to 1.3 billion tonnes annually—is lost or wasted. This represents not only a moral and economic failure but also an inefficient use of natural resources that must be addressed to resolve cross-cutting issues essential for achieving the 2030 Agenda.[1]

The development of active packaging, a technology designed to interact intentionally and positively with food, has emerged as a promising strategy to mitigate food loss and waste, extend shelf life, and improve food safety and quality. Food packaging systems with bioactive components can provide a variety of functions, such as moisture and oxygen absorption, antioxidant effects, free radical scavenging to mitigate oxidation, and inhibition of microbial growth. These effects can either be inherent to the base material of the packaging or introduced by

Received: October 11, 2024
Revised: January 10, 2025
Accepted: January 17, 2025

the addition of bioactive compounds, such as plant extracts and essential oils that functionalize the packaging. By improving the performance of the material, this approach not only extends shelf life and reduces food loss and waste but also promotes sustainability.[2−4]

Active functionalities can be achieved through various application methods, such as using sachets with active ingredients in the packaging, incorporating active compounds into the packaging material, or coating food directly with active suspensions. The incorporation of active components into food packaging systems creates a dynamic, continuous interaction between the packaging, the food, and the external environment. The packaging serves not only as a protective barrier but also as an active system capable of releasing or absorbing certain substances such as moisture, gases, or antioxidants to create suitable internal conditions. At the same time, the food interacts with the packaging through processes such as gas exchange, moisture transfer, or the migration of aromas and compounds that can affect its freshness, safety, and quality. External environmental influences such as temperature, humidity, and oxygen content also influence these interactions and affect both the performance of the packaging and the stability of the food. This synergistic three-way interaction extends the shelf life of the product, inhibits microbial growth, and preserves its sensory and nutritional properties throughout the supply chain—from storage and transportation to marketing and final consumption.[4,5]

Food packaging is crucial for preserving safety and quality, and beyond that it has the potential to advance into active packaging, offering enhanced functionality. However, conventional petroleum-based synthetic plastics, widely used for food packaging, contribute to resource depletion, environmental pollution, and health problems due to their complex and costly recycling process, low biodegradability, and the diffusion of toxic additives and micro- and nanoplastics.[6] To address this challenge, researchers are exploring biodegradable and biobased materials as viable alternatives for packaging solutions that not only align with sustainability goals but also meet growing consumer demand for safe, fresh food and clean labels, as well as functional packaging with minimal environmental impact.[7,8]

In the development of a new generation of food packaging, the focus is on research into environmentally friendly materials from renewable sources. Flexible biobased films, primarily composed of biopolymers such as polysaccharides, proteins, and lipids, provide a sustainable matrix for the incorporation of natural active agents.[4,9,10] Adding essential oil (EO), bioactive compounds, or other functional ingredients to the film-forming suspension (FFS) is a viable way to enhance the antioxidant and antimicrobial properties of biobased films.[3,8] This approach contributes to achieving the desired active properties in food packaging materials.[2] However, EOs are highly volatile and susceptible to oxidation, light, and thermal degradation.[11,12] In addition, when free EO is mixed directly with a FFS or added in the form of conventional emulsions, it tends to be rapidly released, which can reduce the antimicrobial or antioxidant efficacy of the packaging over the shelf life of the food product, posing a challenge for its incorporation into the film-forming process.[13,14] To overcome this challenge, recent approaches involve encapsulating these natural compounds with colloid particles before adding them to the FFS.[15] Therefore, the Pickering emulsion (PE), an emulsion stabilized with biopolymer particles, is the most suitable technique. PE is characterized by higher coalescence stability, stronger protection of the encapsulated compound, lower sustained release rate, safety, biodegradability, and biocompatibility.[16,17]

This review aims to provide a comprehensive analysis of Pickering emulsions with essential oils (EOPE) as innovative active additives for sustainable packaging films, highlighting their potential to improve food safety and quality. By integration of a bibliometric assessment, the current research landscape and trends in the field of EOPE-functionalized packaging are contextualized. In this study, the role of essential oils as natural antimicrobial and antioxidant agents, the stabilization mechanisms of Pickering emulsions with different particle stabilizers, and the physicochemical properties and bioactive performance of EOPE-based packaging systems are discussed. The paper briefly outlines some legal and regulatory considerations for innovative packaging solutions, emphasizing compliance with safety regulations. It also provides valuable insights into current research, highlighting the potential of these solutions as sustainable alternatives for active food packaging systems.

2. METHODS

2.1. Search Strategy and Selection Criteria. In order to analyze recently published studies on EOPE, stabilizer particles, and their performance in sustainable functionalized food packaging films, bibliometric analysis was carried out. The data for this research were collected on March 23, 2024, from the Web of Science (WoS) and Scopus databases.[18,19] The searches were conducted using the following keywords: ("Bio-based film" OR "Biobased film" OR "Active film" OR "Biopolymer" OR "Sustainable packaging" OR "Biodegradable film" OR "Active packaging film" OR "Edible film" OR "Bioactive film") AND ("Essential oil" OR "Essential-oil") AND ("Pickering Emulsion"). Initially, articles published in the last 5 years, from 2020 to 2024 up to the research date, were selected. The articles were then screened by title and abstract, followed by full-text analysis, to identify which studies should be included in the bibliometric analysis. To this end, exclusion criteria comprised undergraduate works, dissertations, theses, and works presented in congresses or symposia, as well as those included in Annals. In addition, studies in languages other than English, reviews, and case studies such as economic evaluations, studies that did not use essential oils, studies that did not use EO to develop PE, or studies that developed only coatings instead of films were also excluded. Thus, the articles selected for this bibliometric study included (a) original research articles and (b) research that focused on the addition of PE based on EO in the formulation of sustainable films.

3. RESULTS AND DISCUSSION

3.1. Pickering Emulsions Containing Essential Oils as Active Additives for Sustainable Packaging Film: Contextualization and Bibliometric Assessment. Incorporating Pickering emulsions containing essential oils (EOPE) with core−shell structures can be a feasible alternative to produce active food packaging films.[20] These films can be used to release bioactive compounds in a controlled manner, enhancing antimicrobial and antioxidant properties to fulfill the requirements of food packaging systems for storage, transportation, and preservation.[21]

The Web of Science (WoS) database identified 85 articles from which 47 articles were selected. Similarly, the Scopus

https://doi.org/10.1021/acsomega.4c09320
ACS Omega XXXX, XXX, XXX−XXX

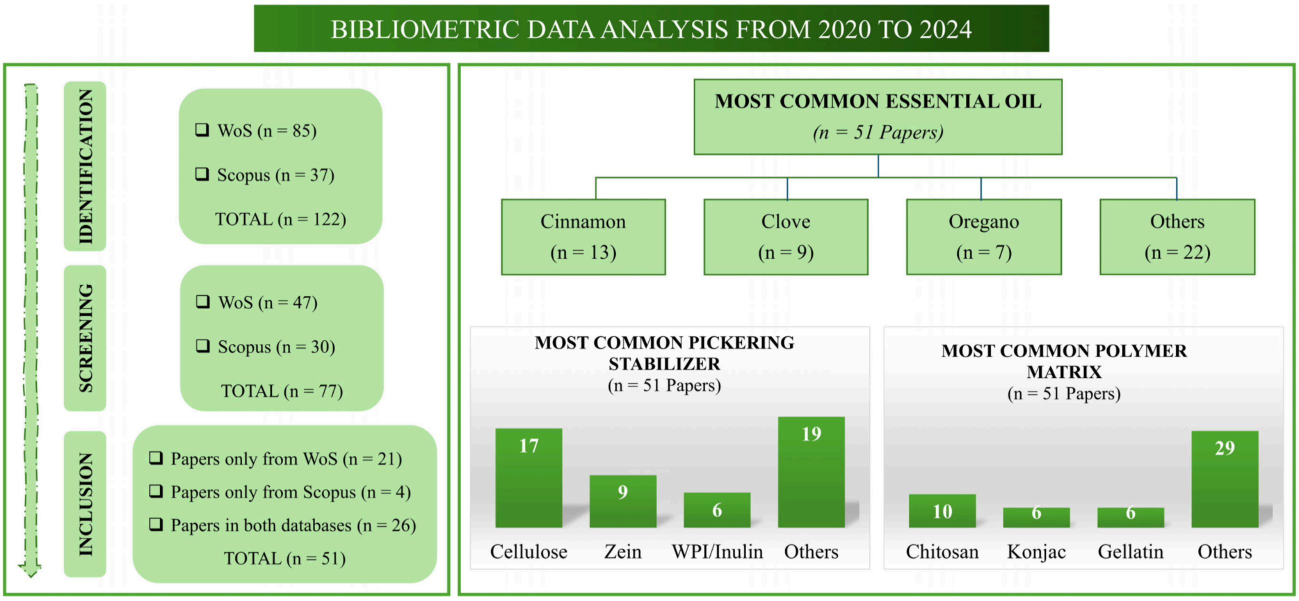


Figure 1. Schematic overview of bibliometric data on the addition of Pickering emulsions based on essential oils in the formulation of sustainable films.

database identified 37 articles, of which 30 were selected. After the selection criteria were applied, 51 articles, excluding duplicates, from 122 records in WoS and Scopus databases were included in the bibliometric analysis. Among the 51 articles published over the last five years and included in the bibliometric data combination, cinnamon (13) was the most studied EO, followed by clove (9) and oregano (7). In addition, the most used solid particle Pickering stabilizer was cellulose (17), followed by zein (6) and the whey protein isolate (WPI)-inulin complex (6). On the other hand, chitosan (10), konjac (6), and gelatin (6) were the most used polymer matrices to which EOPE was added (Figure 1).

3.2. Essential Oils and Their Potential Use for Food Safety and Quality Assurance. EOs are concentrated, volatile, hydrophobic liquids derived from various parts of aromatic plants or fruits as secondary metabolites.[22,23] These parts include roots, stems, seeds, bark, leaves, and flowers.[22,24] Chemically, EOs are a complex mixture of secondary metabolites such as monoterpenes, terpenes, terpenoids, aliphatic compounds, alkaloids, isoflavones, flavonoids, phenolic acids, aldehydes, and carotenoids[23,25] with potent antimicrobial properties against foodborne pathogens such as *Salmonella* sp, *Listeria* sp., and *Escherichia coli* (*E. coli*).[26−28] A possible mechanism of action is that these metabolites target protein groups in the bacterial membrane, altering its permeability and causing bacterial death.[29]

The United States Food and Drug Administration has classified most natural EOs and their extracts as Generally Recognized as Safe or GRAS due to their nontoxic characteristics and safety.[30] Therefore, due to their strong, wide-spectrum activity against microorganisms, EOs are widely used in the food industry as natural preservatives to increase the shelf life of food products such as meat, fruits, vegetables, and dairy.[31] Examples of EOs with antimicrobial and antioxidant activities are listed in Table 1.

3.3. Pickering Emulsions and Particle Stabilizers. Emulsions are colloidal systems composed of at least two immiscible fluids, with one dispersed in the other as small droplets.[16,17] Traditionally, emulsion systems have been composed of oil and water, which can form three types of emulsions: oil-in-water (O/W), water-in-oil (W/O), and complex or multiple emulsions, with O/W emulsions being the most common.[51] However, due to the high surface energy between the two immiscible phases, emulsion systems are considered thermodynamically unstable and susceptible to coalescence over time. Therefore, to ensure that the emulsion remains stable, a surface-active agent or stabilizer such as a chemical surfactant is needed.[52] Most of the surfactants commonly used in traditional emulsions, such as hexadecyltrimethylammonium bromide, benzalkonium chloride, alkylbenzene linear sulfonate,[53] Tween 20, Tween 80, Span 20, and Span 80,[54] present a considerable environmental challenge, particularly in terms of soil and water pollution. Their nondegradability in the environment gives rise to considerable concerns regarding sustainability and the health of ecosystems. Additionally, the potential adverse health effects of these emulsifiers are dependent on the dosage required to stabilize the emulsion.[53,55] As demand for more sustainable systems increases, so does interest in using natural and clean-label ingredients instead of synthetic surfactants.[56]

PEs are stabilized by solid particles (nano- and microparticles) and have been demonstrated to have no toxic effects, particularly when developed using food-grade biopolymer particles,[14,15] lower cost, and easier recovery properties compared to conventional surfactants (Figure 2). Solid colloidal particles can replace surfactants to stabilize oil/water interfaces.[52,56,57] One significant advantage of using these particles is that they can irreversibly adsorb to an oil/water interface and prevent droplet coalescence. This property means that once a particle is at an interface, it will not detach from it spontaneously, even if there are temperature changes. This outstanding stability and encapsulation efficiency are highly desirable and make PE a suitable technology for the protection and controlled release of bioactive compounds that are fixed in a given matrix.[12,58]

Currently, there are several ongoing research studies on the manufacturing strategy of food-grade particles that can stabilize PE systems.[59] To be feasible as a PE stabilizer, the particle

https://doi.org/10.1021/acsomega.4c09320
ACS Omega XXXX, XXX, XXX−XXX

Table 1. Examples of Major Antimicrobial and Antioxidant Activity of Selected Essential Oils

Essential Oil	Plant	Part of the plant used for extraction	Antimicrobial Activity (AA)	Antioxidant Activity[a]	Reference
Anise	*Pimpinella anisum*	Seeds	AA against *Trichoderma viride*, *Penicillium citrinum*, *Aspergillus niger*, *Candida albicans*, and *Staphylococcus aureus* (*S. aureus*)	Antioxidant activity of 20.42% against DPPH assay; 16.49% of radical-scavenging capacity (ABTS) method and total phenolic content of 386.37 μg of GAE/Ml	28, 32
Basil	*Ocimum basilicum*	Leaves	AA against *Brochothrix thermosphacta*, *Carnobacterium maltaromaticum*, *Enterococcus faecalis*, *Staphylococcus xylosus*, *Staphylococcus saprophyticus*, *Listeria innocua*, *Streptococcus salivarius*, *Serratia proteamaculans*, and *E. coli*	Effective antioxidant activity with an IC_{50} of 23.44 ± 0.9 μg/mL compared with Trolox (IC_{50} of 2.7 ± 0.5 μg/mL)	27, 33
Black pepper	*Piper nigrum*	Fruits	AA against *Aspergillus flavus*	Antioxidant activity assessed by β-carotene bleach assay showed lipid peroxidation of 73.6%, higher than that of ascorbic acid (15.18%)	34
Cinnamon	*Cinnamomum verum*	Leaves and bark	AA against *Trichoderma viride*, *Penicillium citrinum*, *Aspergillus niger*, *S. aureus*, *Listeria monocytogenes*, *E. coli*, *Salmonella typhimurium*, and *Salmonella* spp.	Antioxidant activity assessed by β-carotene bleach assay showed lipid peroxidation of 82.3% when compared to synthetic antioxidants, 2,6-di*tert*-butyl-4-methylphenol (BHT) and beta hydroxy acid (BHA) with 74.4 and 81.2%, respectively	26, 35
Citronella	*Cymbopogon nardus*	Leaves	AA against *Candida albicans*, *S. aureus*, *Staphylococcus epidermidis*, *Streptococcus mutans*, and *Salmonella typhimurium*	Mild antioxidant activity with an IC_{50} of 274 ± 2 mg/g compared to BHT and ascorbic acid (IC_{50} of 8029 ± 1 mg/g and 5137 ± 4 mg/g, respectively)	36, 37
Clove	*Eugenia caryophyllata*	Flowers	AA against *S. aureus* and *E. coli*	DPPH scavenging activity IC_{50} ranged from 15.80 to 108.85 μg/mL, depending on flowering stages	38, 39
Ginger	*Zingiber officinale*	Root	AA against *S. aureus* and *E. coli*	Antioxidant activity assessed by β-carotene bleach assay showed lipid peroxidation of 66,5%, lower antioxidant activity than that shown by BHT and BHA (74.4 and 81.2%, respectively)	[35,40]
Grapefruit	*Citrus paradisi*	Peel	AA against *E. coli* and *Leuconostoc mesenteroides*	Antioxidant activity assessed by ABTS and DPPH showed 24 mg Trolox Equivalent (TR)/ml CEO and 15 mg TR/ml CEO, respectively	41
Lavender	*Lavandula angustifolia*	Flowers	AA against *Candida tropicalis* and *Pseudomonas aeruginosa*	Moderate antioxidant activity of 29.08 ± 0.99% of DPPH scavenging capacity	42
Lemon	*Citrus limon*	Peel	AA against *E. coli* and *Leuconostoc mesenteroides*	Antioxidant activity assessed by ABTS and DPPH showed 7 mg TR/ml CEO and 23 mg TR/ml CEO, respectively	41
Lemongrass	*Cymbopogon citratus*	Leaves	AA against *Candida krusei*	Antioxidant activity of 853.0 ± 1.13 μg TR/ml LEO (i.e., 84.0 ± 0.1%) of DPPH scavenging capacity	43
Mandarin	*Citrus reticulata*	Peel	AA against *S. aureus*	Antioxidant activity assessed by ABTS and DPPH showed 31 mg TR/ml EO and 17 mg TR/ml CEO, respectively	41, 44
Marjoram	*Majorana hortensis* Moench	Leaves	AA against *P. aeruginosa*	Mild antioxidant activity with an EC_{50} value of 54.01 mg/mL EO of DPPH scavenging capacity	45, 46
Oregano	*Origanum vulgare* L.	Leaves	AA against *Campylobacter* spp, *Shewanella putrefaciens*, and *Vibrio vulnificus*	Effective antioxidant activity with an EC_{50} value of 7.450 mg/mL EO of DPPH scavenging capacity	46–49
Rosemary	*Rosmarinus officinalis*	Leaves	AA against *Listeria monocytogenes* and *S. aureus*	Moderate antioxidant activity of 28.76 ± 2.68% of DPPH scavenging capacity	42, 50
Thyme	*Thymus vulgaris*	Leaves	AA against *Pseudomonas fluorescence* and *E. coli*	Effective antioxidant activity with an EC_{50} value of 0.944 mg/mL EO of DPPH scavenging capacity	45, 46

[a]Abbreviations: DPPH, 2,2-diphenyl-1-picrylhydrazyl free radical-scavenging capacity; ABTS, 2,2′-azinobis-3-ethylbenzothiazoline-6-sulfonic acid; GAE, gallic acid equivalent; IC_{50} or EC_{50}, concentration of extract necessary to neutralize 50% of initial concentration of free radicals.

https://doi.org/10.1021/acsomega.4c09320
ACS Omega XXXX, XXX, XXX−XXX

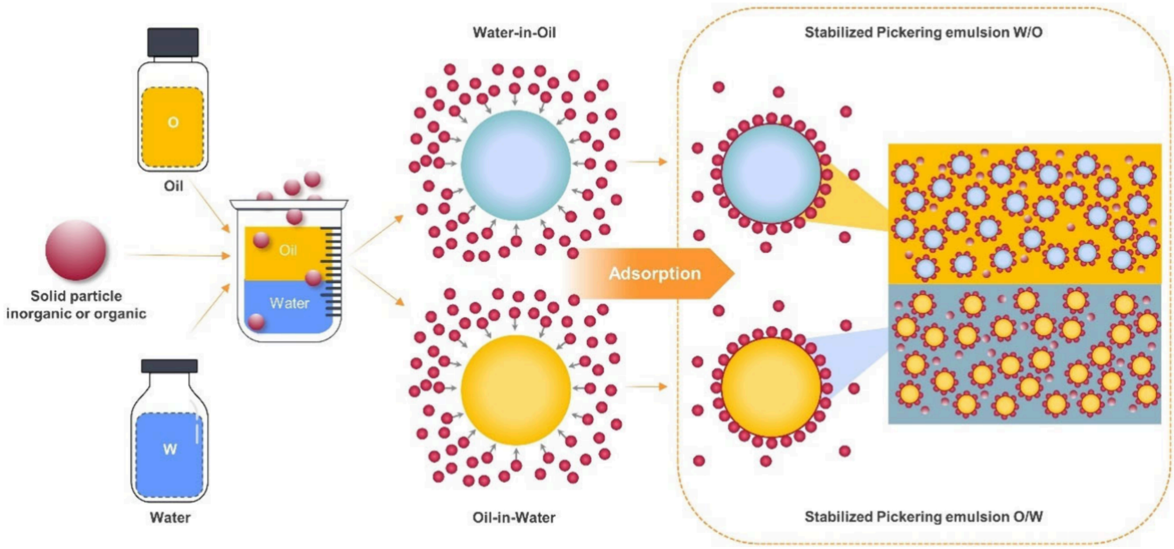


Figure 2. Schematic representation of particle stabilizers in Pickering emulsions.

must be partially wettable by both the continuous and dispersed phases of the system, remain insoluble, present a low surface charge, and be much smaller than the emulsion size.[52] These particles are classified into single and composite particles. Although each solid particle has unique inherent characteristics, such as wettability, size, and shape, which affect the necessary processing conditions differently, single solid particles used as stabilizers for PE can be categorized into three groups based on their origin. These three groups are biological, inorganic, and organic particles. (i) Biological particles are derived from natural biopolymers and are usually renewable, biodegradable, and biocompatible. Examples include cellulose nanocrystals, carboxymethyl chitosan, pea protein nanoparticles, starch, zein, pectin, and gliadin nanoparticles. These particles are particularly attractive for the development of sustainable food packaging due to their functional properties such as film-forming capacity, mechanical reinforcement, antioxidant, and antimicrobial activity. In contrast, (ii) inorganic particles are synthesized from minerals and other nonorganic sources, such as hydrophilically modified silica, halloysite clay nanotubes, kaolinite, and shigaite-like layered double hydroxide particles. These particles offer excellent thermal stability and barrier properties, making them good candidates for improving the structural integrity and shelf life of packaging materials. (iii) Organic particles are usually synthetic polymers such as poly(caprolactone) block-poly-(ethylene oxide) diblock copolymers and oligoimide particles. The choice of the appropriate particle type depends on the specific functional requirements of the packaging system as well as sustainability, regulatory compliance, and end-of-life disposal considerations.[55]

Due to their hydrophilic nature and other inherent properties, certain food-grade biopolymer particles, such as starch, chitosan, and cellulose, exhibit low surface activity at the oil/water interface, which reduces their applicability.[59,60] To overcome such limitations of single particles, composite particles have become a popular area of research, since they generally exhibit superior functionality, particularly in terms of wettability and adsorption behavior, resulting in long-term stability and providing controlled release, making them more useful.[61,62] Composite particle formations can be achieved using proteins, polysaccharides, phenolic compounds, or lipids through solvent mediation and interactions based on the functional properties of biopolymers, particles, and conditions. Examples of composite particles include the following: (I) water-soluble protein-polysaccharide particles, pea protein isolate-high methoxyl pectin and whey protein isolate-dextran; (II) water-insoluble protein-polysaccharide particles, zein-gum arabic and zein-propylene glycol alginate; (III) water-soluble protein-water insoluble polysaccharide particles, soy protein isolate-bacterial cellulose nanofibers, soy protein isolate-chitosan, and pea protein isolate-chitosan; (IV) water-insoluble protein-water insoluble polysaccharide particles: zein-chitosan and gliadin-chitosan.[63]

3.4. Properties and Applications of Pickering Emulsions Containing Essential Oils Functionalized Packaging. Among the 51 papers included in this bibliometric analysis published in the last five years, cinnamon, clove, and oregano EO were the most studied, with 13, 9, and 7 studies, respectively. The growing interest in cinnamon and clove oils is due to their remarkable bioactive properties, which make them attractive candidates for use in active food packaging. Both oils are recognized as safe food additives and are known for their strong antioxidant capacity and broad-spectrum antimicrobial activity against a variety of food-borne microorganisms. Their classification as "Generally Recognized as Safe" (GRAS) increases their appeal as a natural alternative to synthetic preservatives, responding to consumer demand for environmentally friendly packaging solutions.[4,65,67,68] The incorporation of essential oils such as cinnamon, clove, and oregano into packaging films has the potential to improve both bioactivity and functional properties. These oils can contribute to improved mechanical strength and reduced water vapor permeability, which could help protect food by limiting moisture transfer and microbial contamination.[4,64,68,69]

The incorporation of PE based on cinnamon EO (CEO) into collagen films, coupled with oxidized mulberry extract (OME) as a functional enhancer, significantly improved the mechanical and barrier properties of the films. The tensile strength increased from 79.18 to 106.35 MPa, which increased the durability of the films, while the water vapor permeability decreased from 2.82 to 2.30 × 10^{-11} g/(m s Pa) and the water

https://doi.org/10.1021/acsomega.4c09320
ACS Omega XXXX, XXX, XXX−XXX

absorption was reduced from 524.57% to 201.28%, reflecting better moisture resistance. The films also exhibited improved light-blocking, antioxidant, and antimicrobial properties and effectively inhibited *Escherichia coli* and *Pseudomonas fluorescens*. Applied to fish fillets, the CEO PE/OME film extended the shelf life of the product by 4 days and preserved its quality and safety. The film also served as a freshness indicator: it changed color from red on day 0 to black-green on day 12 ($\Delta E > 5$), allowing a visual assessment of the freshness of the fish. These results underline the potential of CEO-based Pickering emulsion collagen films as multifunctional packaging materials that combine longer food preservation with real-time freshness monitoring.[4]

Wu et al.[64] reported that cinnamon EOPE was incorporated into films made from chayote tuber starch, resulting in significant changes in film properties. As the concentration of cinnamon EOPE increased, the tensile strength (TS) decreased from 5.44 to 2.59 MPa, while the elongation at break (EB) increased, reaching 70.8% at 4% EOPE. This decrease in TS is due to the disruption of the structural integrity of the film, which impaired the formation of hydrogen bonds between the starch molecules. The increase in the EB is probably due to the plasticizing effect of the free cinnamon EO, which increases the flexibility of the polymer chains. The water resistance of the films improved with the addition of cinnamon EOPE, especially at concentrations below 2%. This improvement was attributed to the formation of hydrogen bonds between the hydrophilic starch matrix and the hydrophobic particles. The moisture content (MC) of the films decreased slightly from 21.20% to 19.49% due to the hydrophobic properties of cinnamon-EO. However, at higher concentrations (3% and 4%), a slight increase in water solubility was observed, which is probably due to the leaching of the EO. The water vapor permeability (WVP) of the films was reduced by the addition of cinnamon EOPE, with the lowest value (1.24×10^{-10} g/(m s Pa)) observed at 2%. This was attributed to the increased resistance to the movement of water molecules due to changes in the internal structure of the film. These results suggest that cinnamon EOPE can significantly improve the functional properties of starch-based films, contributing to their effectiveness in food packaging.

Yao et al.[66] investigated the effects of cinnamon EOPE stabilized with zein and carboxymethyl tamarind gum on the properties of hydroxypropyl methylcellulose films, also examining the influence of the degree of carboxymethylation. The influence of packaging on the shelf life of cherry tomatoes was also investigated. The droplet size of cinnamon-EOPE decreased significantly from about 93.03 to 10.59 μm as the degree of substitution of carboxymethyl-tamarind gum increased, which promoted a more uniform distribution of droplets in the film matrix. The incorporation of cinnamon EOPE into the hydroxypropyl methylcellulose films resulted in a significant increase in TS from 8.46 to 25.41 MPa, while the water vapor permeability decreased from 6.18×10^{-10} to 4.24×10^{-10} g/(m s Pa), indicating improved barrier properties. The films also showed improved UV protection without compromising transparency, making them ideal for use in food packaging. The films enriched with cinnamon EOPE also showed antibacterial activity against *Escherichia coli* and *Staphylococcus aureus* compared to pure HPMC films. The antioxidant activity was also significantly increased. The EOPE-added films helped to reduce the weight loss of cherry tomatoes compared with unpackaged control tomatoes while slowing the decrease in total soluble solids and titratable acidity, indicating improved preservation. The films contributed to an extended shelf life of the tomatoes by effectively delaying spoilage over a 20 day storage period.

In a study by Zhao et al.,[67] antimicrobial films were produced by incorporating clove EOPE into a matrix of potato starch and poly(vinyl alcohol). The clove EOPE exhibited a zeta potential of −21.7 mV, a droplet size of 186 nm, a polydispersity index of 0.104, and an encapsulation efficiency of 57.9%. These results confirm the successful formation of the emulsion. The clove EOPE was uniformly distributed in the matrix of potato starch and poly(vinyl alcohol), resulting in a smooth and homogeneous film structure. The mechanical properties of the films were significantly affected by the hydrogen bonding and electrostatic interactions between the emulsion and the matrix, resulting in a decrease in TS from 22.4 to 6.80 MPa and a decrease in EB from 375.3% to 91.6%. The films were used for the preservation of pork and were found to provide an extended preservation time of 6 to 10 days and exhibit greater inhibition of *Escherichia coli* compared to *Staphylococcus aureus*. The incorporation of clove EOPE into the films showed a significant antimicrobial effect and potential to improve the preservation of pork.

Bangar et al.[68] explored the potential of cellulose nanocrystals (CNCs) derived from kudzu (*Pueraria montana*) vine, combined with clove essential oil Pickering emulsions (EOPE), to enhance the properties of starch-based films. The incorporation of clove EOPE into a composite film of pearl millet starch and kudzu CNCs led to significant improvements in mechanical properties, with TS increasing from 4.02 to 16.2 MPa, Young's modulus rising from 84 to 398 MPa, and EB decreasing from 48.9% to 30.4%. Moreover, the composite film demonstrated excellent antimicrobial activity against *Staphylococcus aureus* and *Escherichia coli* and was highly effective in extending the shelf life of red grapes, maintaining freshness for up to 15 days at 5 °C.

In another study,[69] the development of films based on konjac glucomannan activated by oregano EOPE stabilized with zein-pectin nanoparticles was investigated. The results showed that the hydrophilicity and hydrophobicity of the konjac glucomannan films can be modulated by adjusting the oregano-EOPE concentration, which has a significant effect on the mechanical and barrier properties of the films. The films with oregano-EOPE to konjac glucomannan ratios of 50:50 and 60:40 exhibited the highest tensile strength (30.98 MPa) and water contact angle (93.56°), respectively, indicating better mechanical properties and higher hydrophobicity. The films showed an effective slow release of the EO for up to 21 days, which underlines their potential for food preservation. The water vapor permeability of the pure konjac-glucomannan films was 5.30×10^{-1} g/(m s Pa), while the oregano-EOPE-added films, especially those containing 60% oregano-EOPE content, showed a significant decrease in WVP to 1.16×10^{-1} g/(m s Pa), indicating an improved water vapor barrier. Increasing the oregano EOPE concentration from 0% to 60% resulted in a steady decrease in WVP, which was attributed to the formation of a more compact network and a higher crystallinity of the films. The addition of oregano EOPE to konjac-glucomannan films resulted in suitable films for fruit preservation.

Some recently published studies on the properties of EOPE-functionalized films applied to various foods are listed in Table 2.

https://doi.org/10.1021/acsomega.4c09320
ACS Omega XXXX, XXX, XXX−XXX

Table 2. Influence of EOPE Functionalization on Sustainable Food Packaging's Mechanical, Physical, Thermal, Antimicrobial, and Antioxidant Properties[a]

Essential Oil	Polymer Matrix	Pickering Stabilizer	Antimicrobial effect	Antioxidant Activity	Food Application	Reference
			Animal-Source Foods			
Alpinia galanga	PVA/acetylated pullulan	Soybean protein isolate-chitosan	EOPE contributed to the antibacterial activity and showed a dose-dependent effect; moreover, the antibacterial effect on *S. aureus* was more effective than that on *E. coli*	The inclusion of EOPE demonstrated a strong DPPH and ABTS scavenging ability, and the strength of the antioxidant ability showed a dose dependence with the addition of the emulsion	Extend chicken's shelf life and maintain sensory parameters such as pH and color	70
Cinnamon	Chayote tuber starch (CTS)	Zein-pectin nanoparticle	Composite films incorporated with at least 2% of EOPE showed stronger antibacterial activity against *S. aureus* and *E. coli*	The DPPH radical scavenging of the films increased significantly from 9.51% to 56.92% as the EOPE content increased from 0% to 4%, respectively	The addition of 2% EOPE significantly slows the formation of TVB-N, delaying the degradation of ground beef and extending its shelf life	64
	Chitosan	Cellulose nanocrystal	The films containing EOPE exhibited antimicrobial activity against *E. coli* and *S. aureus*. The inhibition zones of the films increased as the EO content was increased	NA	The preservation of pork pieces was enhanced while maintaining their structural stability	71
	Collagen	Soy protein isolate-chitosan	Inhibiting the growth and reproduction of *E. coli* and *Pseudomonas fluorescens*	DPPH radical scavenging of 50.17% was achieved with the addition of the EOPE	Fish preservation and freshness indicator through pH sensitivity	4
Cinnamon and perilla	Chitosan nanoparticles/Anthocyanidin/Collagen	Collagen	At the end of storage, films containing EOPE exhibited the lowest total bacterial count (5.89 ± 0.01 l g (CFU/g)) compared to the control group (10.85 ± 0.01 l g CFU/g)	NA	Preservation of fresh red sea bream fillets and delayed lipid oxidation and proteolysis. The shelf life of fillets was extended from 6 to 14 days	72
	Anthocyanidin/Chitosan	Collagen	NA	The incorporation of various concentrations of EOPE resulted in greater antioxidant activity, as measured by DPPH scavenging activity, compared to the control group samples (i.e., EOPE 0%)	The freshness of packaged fish fillets was extended in terms of thiobarbituric acid (TBA) levels	73
Clove	Potato starch/polyvinyl alcohol (PVA)	WPI-inulin	The film showed potent inhibition zones against *S. aureus* and *E. coli*. As the EO concentration in PE increased, the diameter of the inhibition zones increased more significantly	The films added with EOPE exhibited great DPPH scavenging activity; however, the additional amount of EOPE did not noticeably alter the antioxidant activity. Therefore, no dose dependence was observed	Extending the preservation period of pork meat from 6 to 10 days	67
	Gelatin/agar	Copper-modified zinc oxide nanoparticles	The EOPE film exhibited antimicrobial activity against both Gram-positive (*Listeria monocytogenes*) and Gram-negative (*E. coli*) bacteria. However, it demonstrated slightly greater antibacterial activity against *L. monocytogenes* than against *E. coli*	The pure gelatin/agar-based film exhibited a modest ~6% DPPH and ~33% ABTS free radical scavenging; however, the incorporation of EOPE increased this free radical scavenging activity to ~33% and ~61% against DPPH and ABTS, respectively	The use of EOPE film to wrap pork belly resulted in delayed bacterial growth, as evidenced by the total aerobic bacterial count (TABC). Even after 8 days, the TABC value of the test group did not exceed 6.7 log CFU/g	74
Grapefruit	Lotus seed drill core powder starch	Corn nanostarch	Adding 20% EOPE exerted antibacterial activity against *E. coli* and *S. aureus*; the inhibition zone diameters of *E. coli* and *S. aureus* increased significantly to 11.71 ± 0.03 mm and 10.25 ± 0.33 mm, respectively	NA	Prolong the shelf life of refrigerated pork meat pieces, based on TVB-N and TBARS reduced content	65
Lavender	Gelatin	Gelatin	The film containing EOPE exhibited antibacterial activity against *S. aureus* and *E. coli*; additionally, the formulation demonstrated controlled release of the EO, indicating potential for long-term preservation	All films developed with EOPE exhibited a high ABTS+ scavenging capacity	Real-time monitoring and maintenance of shrimp freshness	75
Nutmeg	Low-density polyethylene (LDPE)	Inulin-WPI	The interaction between LPDE/PE slowed release of the active ingredients compared to the free/coarse emulsion or the nanoemulsion; the decrease in *E. coli* growth suggests an increase in bacterial inhibition due to the higher concentration of EOPE	The concentration of EOPE significantly increased the DPPH-scavenging activity of the film, with values ranging from 38 to approximately 66%	Preserve the quality and extend the shelf life of Tilapia fish fillets, based on TBARS and TBV-N values	76
Thymus vulgaris	Tapioca starch/polyvinyl alcohol (PVA)	Cellulose nanocrystals	The composite film containing 20% EOPE exhibited bacteriostatic properties for up to 8 days, as determined by the total viable counts (TVC)	NA	Composite film with 20% TEVO PE extends fish fillet freshness for up to 8 days	77
Zataria multiflora	Chitosan	Zein	The microbiological analysis was performed by total viable count (TVC); chitosan coatings with EOPE showed stable and long-term antimicrobial activity compared to the coating with free EO	NA	EOPE delayed chemical and microbial spoilage, extending the shelf life of salmon (*Salmo trutta*) stored at refrigerator temperatures	78

https://doi.org/10.1021/acsomega.4c09320
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Table 2. continued

Essential Oil	Polymer Matrix	Pickering Stabilizer	Antimicrobial effect	Antioxidant Activity	Food Application	Reference
Plant-Source Foods						
Anise	Corn starch/cassia gum	Sodium starch octenyl succinate	EOPE-incorporated composite films showed stronger dose-dependent antibacterial activity against *S. aureus* and *E. coli*	The DPPH radical scavenging ability of the composite films significantly increased by 18.45% with the addition of the EOPE with an oil/water phase ratio of 6:4. A similar improvement was observed in the scavenging ability of the composite membrane against ABTS radicals	Delay the browning and spoilage and maintain the original quality of *Agaricus bisporus* (mushroom)	79
Basil	Corn starch	Cellulose nanofiber	NA	NA	Maintain quality and prolong the shelf life of mandarin oranges	80
Cinnamon	Hydroxypropyl methylcellulose (HPMC)	Zein-carboxymethyl tamarind gum	Films containing EOPE showed stronger antimicrobial activities against *E. coli* and *S. aureus*	The addition of EOPE to the film matrix increased the DPPH scavenging activity by 52.00%, indicating its antioxidant ability	Increasing the shelf life of cherry tomatoes	66
Clove	Pearl millet starch/kudzu (*Pueraria montana*) cellulosic nanocrystals (CNC)	Kudzu CNC	Films with EOPE exhibited remarkable antimicrobial properties against *S. aureus* and *E. coli*	NA	Composite films containing EOPE were found to be highly effective in extending the shelf life of red grapes up to 15 days at 5 °C; the films maintained the weight, firmness, and soluble solids of the grapes	68
Ginger	Carboxymethyl cellulose/polyvinyl alcohol (PVOH)	Oleic acid	The EOPE-incorporated films showed antimicrobial effects that increased with increasing concentration of EOPE; the 3% EOPE film showed the highest antifungal activity against *Penicillium digitatum*	The incorporation of EOPE led to a dose-dependent increase in polyphenolic compounds in the films; furthermore, the films with added EOPE (0.5−3%) exhibited significantly higher antioxidant activity in DPPH scavenging effects (10.16%, 17.16%, and 21.09%, respectively)	Increasing the shelf life of bread slices from 4 to 30 days	81
Lemongrass	Chitosan	Cellulose nanofibers	The addition of EOPE to the film suppressed the fungal activity of *Botrytis cinerea* in inoculated tomatoes in an in vivo antifungal test	NA	The controlled weight loss and fungal contamination during storage increased the shelf life of tomatoes	82
Litsea cubeba	Gelatin/ZnO nanoparticles	Pectin-β-cyclodextrin	The film exhibited remarkable antimicrobial properties against *Colletotrichum gloeosporioides* and *Listeria monocytogenes*	The addition of EOPE to the film matrix resulted in up to 73% DPPH radical scavenging activity	The optimized films improved the postharvest quality and reduced the anthracnose of fresh mango samples during storage	51
Oregano	Konjac glucomannan	Zein−pectin nanoparticle	The EOPE-functionalized film resulted in significant antibacterial effects, inhibiting the growth of *E. coli* at the initial stage more effectively than that of *S. aureus*	The film's oxidation resistance was found to increase with a higher concentration of EOPE. In the DPPH assay system, the 75% EOPE film exhibited the highest DPPH radical scavenging activity and an antioxidant capacity comparable to that of vitamin C.	Strawberries wrapped in films showed softening instead of spoilage, confirming the films' ability to preserve freshness	69

[a]NA: not assessed.

https://doi.org/10.1021/acsomega.4c09320
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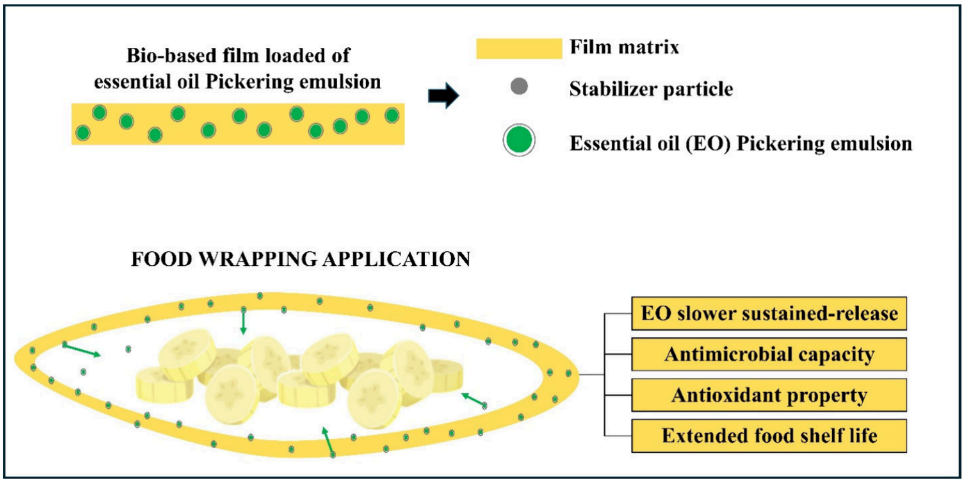

Figure 3. Schematic representation of the advantages of using biobased packaging for food with added essential oil by Pickering emulsion.

As Table 2 shows, EOs are interesting candidates for use as natural antioxidant and antimicrobial additives for the functionalization of food packaging.[30,83] However, they are usually highly sensitive to oxidative conditions and elevated temperatures, which can trigger chemical instability, volatilization, oxidation, and susceptibility to degradation,[13,31,84] thereby posing several challenges for their incorporation into the film-forming process.[69,85] However, incorporating EO into biobased films through PE has been shown to enhance their stability while providing multiple functional benefits, including improved antioxidant capacity, antimicrobial activity, and extended shelf life of packaged products (Figure 3).

Ongoing research continues to investigate various methods for developing food-grade particles that stabilize Pickering emulsions. From the recent studies in Table 2, individual particles such as corn nanostarch,[65] cellulose nanocrystals[71,77] or nanofibers,[80,82] zein[78] and collagen[72,73] are more commonly used as PE stabilizers, although the benefits of composite particles have been reported in the literature. Some work has shown that the combination of polysaccharides and proteins is more useful, as they generally have higher long-term stability.[59,60]

Polysaccharides generally have hydrophilic properties and, therefore, have limited surface activity at the oil−water interface. An effective strategy to solve this problem is the combination of polysaccharides and proteins, which optimizes the behavior at the interface by balancing their hydrophilic and hydrophobic properties, thus increasing the stability and functionality of emulsified systems.[59,60] On the other hand, encapsulation by the PE technique may reduce the antibacterial activity of the EOs to a certain extent due to the controlled release of the encapsulated active ingredients. However, this controlled release may also help to maintain functional activity during long-term storage.[20,64]

Li et al.[59] modified pea protein and chitosan to produce composite particles that effectively stabilize a Pickering emulsion with a high-volume fraction of oil phase (75% corn oil). The incorporation of polysaccharides such as chitosan into proteins such as pea protein significantly improved the emulsifying properties and resulted in a stable emulsion with a low susceptibility to coalescence and phase separation. This stability is attributed to the strong interactions and steric repulsion by the polysaccharide-protein complexes at the oil−water interface, which improves the structural integrity of the emulsion. The optimized stability of these emulsions allows them to be used as effective fat substitutes in various foods, such as pork sausages, without compromising texture or sensory properties.

In addition, the use of such emulsion systems allows the encapsulation and controlled release of bioactive compounds, which improves their bioavailability and chemical stability during processing and storage.[60] This dual function of improving emulsion stability and providing nutrients makes Pickering emulsions of polysaccharides and proteins highly beneficial for the development of healthier foods with improved quality characteristics. The blending of polysaccharides and proteins in Pickering emulsions is therefore a promising strategy for innovative food packaging and formulation solutions.[60]

3.5. Legal Requirements for the Market Entry of Innovative Packaging Concepts Containing Pickering Emulsion. Due to their antioxidant and antimicrobial potential, the PE described in this paper are promising resources for the design of innovative active packaging concepts. However, before an active packaging concept can be successfully introduced to the market, it must be safe and comply with regulatory requirements.[86] In Europe, the central regulation governing materials that come into direct contact with food is regulation (EC) No. 1935/2004. This regulation aims to oversee materials such as packaging that directly touch food and their introduction to the market while also ensuring the protection of human health (Reg (EC) No. 1935/2004). The European Union allows the use of active packaging as long as it is safe, effective, and complies with all the requirements of regulations No. 1935/2004 and No. 450/2009, which concerns "active and intelligent materials and articles intended to come into contact with food" (Reg (EC) No. 450/2009).

As described in this paper, nanoparticles can be used as stabilizers for PE. The use of substances in nanoform is not explicitly mentioned in regulation (EC) No. 1935/2004. Nevertheless, regulation (EU) No. 10/2011, which specifically regulates plastic materials in direct contact with food, allows their use as long as they are listed in and used according to a positive list provided within the regulation (Reg (EU) No. 10/2011). Nanoform substances have different chemical and physical properties compared to their conventional counterparts, mainly due to their reduced particle size, larger surface area, and higher reactivity. Recognizing these differences, Reg

https://doi.org/10.1021/acsomega.4c09320
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(EU) No. 10/2011 requires all nanoscale substances to undergo a case-by-case risk assessment by the European Food Safety Authority (EFSA). This tailored assessment ensures a comprehensive understanding of the unique toxicological profiles associated with nanomaterials before they are approved for food contact applications. The assessment process includes several crucial steps: (i) a detailed characterization of physicochemical properties, including particle size, morphology, and surface chemistry; (ii) a toxicological evaluation to assess potential mutagenic, carcinogenic, and reproductive toxicity; (iii) migration testing to quantify the transfer of nanoparticles from the packaging into the food matrix; (iv) an exposure assessment to establish safe consumption levels. Only nanoparticles that are classified as safe after this rigorous assessment will be included in the Union list of authorized substances, subject to certain conditions for their use.

4. CONCLUSION AND PERSPECTIVES

Pickering emulsions, stabilized by various solid particles and macromolecules, have emerged as a promising alternative for incorporating and ensuring the sustained release of essential oils into biobased polymer matrices.

To ensure the integrity of the film during storage and transportation and to prevent the growth of microorganisms, an effective approach is to increase the microstructural density of the films by improving the polymer matrix cross-linking. This improvement can be accomplished through physical, chemical, or enzymatic methods. Chemical modifications involve the use of additives to cross-link the polymer chains. Enzymatic modifications catalyze the binding of natural mediators to the side chain groups of the polymer matrix molecules.[4] Alternatively, low-pressure plasma (LPP) provides a nonthermal option for enhancing the adhesion of active ingredients and the physical properties of packaging materials. Treatment with LPP has been shown to improve material properties by introducing new functional groups on the surface, thereby enhancing mechanical and hydrophilic properties.[76] Therefore, biobased food packaging systems functionalized with EOPE show significant potential for improving food preservation. These systems enhance antimicrobial activity against common food contaminants, including *Listeria monocytogenes*, *S. aureus*, and *E. coli*. They also exhibit antioxidant properties, including ABTS and DPPH radical scavenging abilities. Additionally, they promote sustainability by reducing the reliance on synthetic materials and effectively extending the shelf life of packaged foods.

The successful implementation of EOPE-based packaging in commercial applications requires a multidisciplinary approach that addresses critical challenges such as scalability, regulatory compliance, and environmental impact. To bring EOPE film production from the laboratory to industry, formulation and processing techniques must be optimized to ensure cost efficiency and compatibility with existing packaging machinery. In addition, the development of standardized protocols to assess the migration of active ingredients and nanoparticles in food matrices is essential to ensuring consumer safety and regulatory approval. Collaboration with regulatory authorities and industry representatives can facilitate the creation of safety guidelines and speed up the commercialization process.

It is anticipated that the commercial use of antimicrobial and antioxidant materials derived from natural resources will increase, leading to enhanced safety and longer shelf life. Thus, future research endeavors should prioritize the incorporation of intelligent systems with active compounds through the utilization of nanotechnological methodologies. This approach may prove to be an effective means of reducing the potential negative impact on the polymer matrix structure and mechanical properties, especially in the case of biopolymer-based matrices.

Equally important is the assessment of the environmental footprint of EOPE-based packaging through comprehensive life cycle assessments (LCA). Such assessments can help identify opportunities for improvement, such as improving the biodegradability of the films and integrating circular economy principles into their design and disposal. Consumer acceptance studies and market analysis are also critical to understanding the demand for these sustainable packaging solutions and matching their characteristics to consumer expectations.

The ongoing advancements in biotechnology, analytical chemistry, microelectronics, and materials science offer the potential to create innovative, intelligent packaging solutions. Furthermore, the migration of packaging constituents, particularly in the case of nanoparticles, must be subjected to rigorous toxicological analysis and evaluated against the established regulatory limits to ascertain their long-term safety for human health. These developments have the potential to assist in achieving industrial standards for food safety and minimizing the environmental impact, thereby facilitating broader applications of active packaging in the food industry.

■ ASSOCIATED CONTENT

Data Availability Statement

Data not shared.

■ AUTHOR INFORMATION

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https://doi.org/10.1021/acsomega.4c09320
ACS Omega XXXX, XXX, XXX−XXX

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Funding

This research work was funded and supported by the Research Support Foundation of the State of Rio de Janeiro (FAPERJ) (code E-26/010/002738/2019; code E-26/211.985/2021; code E-26/201.428/2022), the São Paulo Research Foundation (FAPESP, Brazil) for the Young Investigator grant (2020/08727-0 and 2021/05947-2). The Article Processing Charge for the publication of this research was funded by the Coordination for the Improvement of Higher Education Personnel - CAPES (ROR identifier: 00x0ma614).

Notes

The authors declare no competing financial interest.

ACKNOWLEDGMENTS

The authors would like to thank the staff and facilities of the Rio de Janeiro State University (UERJ), the Federal University of the State of Rio de Janeiro (UNIRIO), the University of São Paulo (USP), and the Sustainable Packaging Institute (SPI) of Albstadt-Sigmaringen University.

ABBREVIATIONS USED

ABTS 2,2′-azinobis-3-ethylbenzothiazoline-6-sulfonic acid
AFM atomic force microscope
BHA beta hydroxy acid
BHT 2,6-di-*tert*-butyl-4-methylphenol
CFU/g colony-forming unit per gram
DPPH 2,2-diphenyl-1-picrylhydrazyl
E. coli *Escherichia coli*
EB elongation at break
EC European commission
EO essential oil
EOPE essential oil Pickering emulsions
FFS film-forming suspension
GAE gallic acid equivalent
GRAS Generally Recognized as Safe
HPMC hydroxypropyl methylcellulose
LPDE low-density polyethylene
O/W oil-in-water emulsion
PE Pickering emulsion
PVA poly(vinyl alcohol)
S. aureus *Staphylococcus aureus*
SEM scanning electron microscope
TBARS thiobarbituric acid reactive substances
TS tensile strength
TVB-N total volatile base nitrogen
TVC total viable counts
W/O water-in-oil emulsion
W/O/W water-in-oil-in-water emulsion
WoS Web of Science
WPI whey protein isolate
WVP vapor transmission

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https://doi.org/10.1021/acsomega.4c09320
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