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Effect of Nanoparticle and Essential oil Addition on Kappa Carragenan-based Bionanocomposite Films: A Review

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Authors' contributions

This work was carried out in collaboration among all authors. All authors read and approved the final manuscript.

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Review Article

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ABSTRACT

The issue of plastic waste has become a global concern due to its negative impact on environmental sustainability. Bionanocompsites are a combination of composite materials made from natural materials with nano-sized organic or inorganic fillers that are biodegradable. Kappa carrageenan-based bionanocomposite films offer an eco-friendly solution for packaging. Kappa carrageenan is one of the natural polymers that can be used as a biofilm constituent material which is recognized to form a good film due to its ability to form a strong and stiff gel, but still has limitations in water vapor permeability and lower mechanical properties. The use of nanoparticles and essential oil into carrageenan-based films can improve the characteristics including mechanical properties, optical properties, barrier properties, and act as antibacterial and antioxidant in the films.

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Keywords: Nanoparticles; bionanocomposite film; essential oil; kappa carrageenan.

1. INTRODUCTION

The issue of plastics is a global concern with a drastic increase in production over the last few decades [1]. The high use of plastics has a negative impact on environmental sustainability [2]. Petroleum-based synthetic polymers have been widely used in various types of plastic packaging, but are a major source of waste problems [3]. Petroleum polymer-based plastics are difficult to degrade, require tens to hundreds of years to disintegrate and if burned, plastics will produce carbon emissions that pollute the environment [4,5]. Because of these weaknesses, a plastic packaging technology solution that is easily degradable and environmentally friendly is needed.

Several studies have led to technologies to make plastics from rapidly degradable natural materials, known as biodegradable plastics or bioplastics. Bionanocomposutes are attracting great attention as potential replacements for petroleum-based plastics as they can extend the shelf life of food products serving as barriers against the transfer of water vapor, gases, lipids, and flavor components [6].

These bionanocomposites are made from biopolymers (natural polymers) such as starch, cellulose, and fats [7]. Biopolymers are polymeric materials obtained from natural resources (animals, plants, and algae). Commonly used biopolymers in the formulation of bionanocomposite films are polysaccharides, proteins, and lipids [8]. Among those biopolymers, mainly seaweed-based polysaccharides (alginate and carrageenan) have received great attention in recent years due to their good barrier properties as well as mechanical properties [9].

Processing carrageenan into biofilm is expected to boost the growth of the carrageenan industry. Kappa carrageenan is one type of carrageenan that is recognized as a good film-former because it has the ability to form strong and rigid gels with hydrophobic properties towards water [10,11]. However, kappa carrageenan-based films still have limitations in water vapor permeability and lower mechanical properties [12]. One way to improve the mechanical properties and permeability of films from kappa carrageenan is to incorporate nanoparticles as fillers [13]. In addition to nanoparticles, the addition of essential oils can be used as additives in bionanocomposites. Essential oils extracted from plants and spices are proven to provide antioxidant, antimicrobial effects, and reduce water vapor permeability of hydrophilic films [14]. Based on this information, it can be seen that the addition of nanoparticles and essential oils to kappa carrageenan-based bionanocomposite films can improve the characteristics of bionanocomposite films for the better. Further study needed to see the effect of the addition of nanoparticles and essential oils on mechanical properties, optical properties, barrier properties, especially antioxidant active properties and antimicrobial properties.

2. EFFECTS OF EO AND NANOPARTICLES ON MECHANICAL PROPERTIES OF BIONANOCOMPO-SITE FILMS

The mechanical properties of bionanocomposite films include characteristics such as thickness, tensile strength, and elongation. Most studies have reported that the addition of nanoparticles and essential oils has a significant effect on the mechanical properties of bionanocomposite films (Tabel 1). Tensile strength, elongation, and thickness are key factors that help to explain the relationship between the mechanical properties of film materials and their chemical structure [15].

The addition of nanoparticles such as SiO2 and ZnO to the carrageenan biofilm was shown to improve the mechanical properties of the film. This can occur because the bead milling process of these nanoparticles results in better dispersion, reduced particle size, and improved distribution within the biofilm matrix [16]. Based on research [13] stated that the addition of ZnO nanoparticles with an optimal concentration of 0.5% in kappa carrageenan/ZnO-based films increased tensile strength and decreased elongation, but beyond this concentration, tensile strength actually decreased.

Essential oils such as clove oil, tea tree oil, and oregano oil can also improve the flexibility and elongation of bionanocomposite films. Essential oils can interact with polymer chains, thereby increasing the plasticity of the film and its ability to stretch without breaking [17]. The use of essential oils decreases the stiffness and increases the flexibility of the material in the film which contributes to the increased toughness of the biofilm. The combination of using nanoparticles and essential oils together can optimize the mechanical performance of biofilms. Nanoparticles provide the necessary stiffness and strength, while essential oils contribute to flexibility and toughness, resulting in biofilms with balanced and enhanced mechanical properties [18]. This combination results in a bionanocomposite film 80 that is not only strong but also flexible enough for applications that require high stretchability.

In several studies of bionanocomposite films, it was reported that film thickness can be increased by the presence of nanoparticles. The addition of TiO2 nanoparticles to semi- refined carrageenan-based films creates a more complex matrix, which directly increases the thickness of the film [19]. This was added by [20] which states the addition of ZnO nanoparticles as a film admixture acts as a good filler.

This is because nanoparticles have a large surface area and surface energy so that they can build strong interactions and can be well dispersed in the matrix. The addition of nanoparticles can increase the total solid content in the film-forming solution so that the thickness of the film increases [21]. In addition to nanoparticles, the addition of essential oil to the bionanocomposite film significantly increases the film thickness.

The addition of ginger essential oil to chitosan-based films significantly increases the film thickness as the concentration of GEO (Ginger Essential Oil) increases due to the interaction between chitosan and GEO, which reduces the polymer chain arrangement [22]. The incorporation of nanoparticles and essential oil also significantly increases the thickness of bionanocomposite films. This is evidenced in the study [23] that the incorporation of TiO2 nanoparticles with ZEO (Zataria multiflora Essential Oil) increased the thickness significantly to 0.071 and 0.073 mm.

Overall, the addition of nanoparticles and essential oils in bionanocomposite films offers significant improvements to the mechanical and functional properties of the films. The synergy between these two components creates a superior material that can be optimized for various applications including food packaging.

3. EFFECT OF EO AND NANOPARTICLES ON BARRIER PROPERTIES OF BIONANOCOMPOSITE FILMS

Barrier properties are important parameters that determine efficiency in food packaging. Packaging materials must be able to protect food from spoilage and deterioration [21]. The ability of the film to slow down the loss of moisture from the product is an important characteristic that affects product quality [24,25]. The water vapor permeability (WVP) of packaging films is one of the important properties for assessing their suitability as packaging materials as well as an important component for predicting product shelf life [26]. This parameter can be used to estimate the barrier properties of the film by measuring the rate of diffusion of water molecules through the cross-section [27]. Most studies have reported that the addition of nanoparticles and essential oils has a significant effect on the barrier properties of bionanocomposite films (Table 2).

The addition of nanoparticles into the biocomposite film matrix significantly reduces water vapor permeability. The nanoparticles act as a physical barrier that extends the diffusion path of water vapor molecules through the film. In an article [28], the addition of ZnO and TiO_2 nanoparticles in kappa carrageenan-based films has been shown to improve the barrier properties against water vapor. These nanoparticles fill the gaps in the polymer matrix and strengthen the bonds between polymer chains, thereby reducing water vapor permeability. In research [29], the addition of SiO2 nanoparticles in kappa carrageenan-based films also showed a significant reduction in water vapor permeability. This was attributed to the barrier properties of the SiO2 nanoparticles. SiO2 nanoparticles, having a high surface area, will block or trap water vapor molecules [30].

Essential oils are known to have antimicrobial properties and can affect the barrier properties of biocomposite films. In research [31], the addition of Zataria multiflora Boiss (ZEO) and Mentha pulegium (MEO) essential oils into kappa carrageenan films reduced water vapor permeability. Essential oils reduce the density of the film structure, thereby reducing the WVP of the film. Other studies in the research [32] showed that the addition of essential oils into chitosan films also reduced water vapor permeability. Essential oils increase the resistance to water vapor by modifying the microstructure of the film and filling the existing gaps, thereby reducing the diffusion path of water vapor.

The combined use of nanoparticles and essential oils in the formulation of biocomposite films provides a stronger synergistic effect toward reducing water vapor permeability. For example, in a study by [33], the combination of ZnO nanoparticles and cinnamon essential oil in PLA films resulted in a significant reduction in water vapor permeability compared to the use of nanoparticles or essential oil separately. This combination creates a denser and more homogeneous structure, making it more effective in inhibiting water vapor diffusion.

Solubility is another important property that affects the barrier ability of the film. Literature study [13] on kappa carrageenan-based bionanocomposite films showed the addition of ZnO nanoparticles significantly increased the solubility, but the higher concentration of ZnO decreased the solubility. This is because the addition of ZnO nanoparticles to the film increases the formation of hydrogen bonds between ZnO and matrix components, so free water molecules interact less with the film. Similar results were obtained by carrageenan-based bionanocomposite films with the addition of SiO2 nanoparticles, ZnO nanoparticles, and CuO nanoparticles. Showing that the added nanomaterials can reduce the solubility of the film [28].

The addition of essential oils (ZEO and MEO) to carrageenan-based films showed effects on their solubility. Films with Zataria multiflora Boiss (ZEO) and Mentha pulegium (MEO) added significantly decreased the solubility of the films. This is due to the increased interaction between the hydroxyl groups of carrageenan and essential oil components which reduces the availability of hydroxyl groups and water-polymer interactions [31]. This decrease in solubility is beneficial for product integrity and water resistance. Similar results were also found in the research [34] with the addition of Clove essential oil (CEO) to Alginate/kappa-carrageenan film slightly decreased its water solubility. This is due to the hydrophobic nature of CEO which reduces the interaction between film particles and reduces the interaction of polysaccharides with hydroxyl groups, thereby increasing the hydrophobicity of the film and reducing water solubility.

Another parameter that can describe the barrier properties in films is the water contact angle. The water contact angle is usually used to determine the hydrophilicity or hydrophobicity of the film. A study [28] showed that the addition of CuO nanoparticles to carrageenan films increased the water contact angle (WCA) significantly. This was due to increased bonding, nanoparticle equalization, and air trapping in the biopolymer matrix. Similar results were found in the addition of essential oils to the water contact angle of bionanocomposite films. Research [35] showed that increasing the concentration of palm oil can increase the WCA of the film. This indicates that essential oil makes the surface of the bioplastic film more hydrophobic. Similar results were found in the research [36] which showed the incorporation of thyme and clove essential oils into PLA/PBAT bioplastic films at higher concentrations significantly increased the water contact angle. This could potentially improve the suitability of the film for applications that require moisture resistance.

4. EFFECTS OF EO AND NANOPARTICLES ON OPTICAL PROPERTIES OF BIONANOCOMPOSITE FILMS

Optical characteristics are one of the important properties of packaging films as they relate to UV blocking ability which prevents food oxidation and transparency which influences consumer preferences in assessing food freshness [12]. Two optical properties that are often analyzed are UV-screening and transparency properties. UV-screening refers to the film's ability to block ultraviolet radiation, while transparency refers to how much visible light can pass through the film unimpeded [20]. Most studies have reported that the addition of nanoparticles and essential oils has a significant effect on the optical properties of bionanocomposite films (Tabel 3).

The addition of nanoparticles to bionanocomposite films can improve UV-screening properties. In research [37], It was found that films with the addition of SiO2 nanoparticles in kappa-carrageenan films showed a decrease in light transmission and improved UV-screening properties. This is due to the ability of SiO2 nanoparticles to absorb and scatter UV light, thus preventing damage due to UV radiation. The addition of nanoparticles decreases transparency, due to the nature of the polymer and the agglomeration of nanoparticles which affects the light transmission of the film. In addition, research [38] proved that the incorporation of amorphous cellulose

Table 1. Mechanical Properties in Bionanocomposite Films

Polymer Matrix	Nanocomposites and Essential Oils	Thickness (mm)	Tensile Strenght (MPa)	Elongation (%)	References
SR Kappa Carrageenan	SRkC/ZnO (0,5%)	-	32.03 ± 1.1	5.17 ± 1.0	[13]
	SRkC/ZnO (1,0%)		26.40 ± 3.7	5.50 ± 2.6	
	SRkC/ZnO (1,5%)		22.77 ± 0.8	4.70 ± 0.8	
Carrageenan	Car/agar	51.9 ± 5.5	45.4 ± 6.4	2.5 ± 0.8	[17]
	Car/agar/ZnS (2 wt%)	57.8 ± 5.5	60.4 ± 7.2	4.0 ± 1.0	
	Car/agar/PET	53.1 ± 6.5	41.3 ± 9.6	3.7 ± 1.1	
	Car/agar/ZnS (2 wt%)/ PET	56.3 ± 3.7	45.8 ± 9.3	5.3 ± 0.9	
Carrageenan/Gelatin	Ge/Car	0.304 ± 0.01	14.42 ± 0.5	19.59 ± 0.8	[18]
	Ge/Car/TEO (10%)	0.425 ± 0.01	12.28 ± 0.7	45.21 ± 1.0	
	Ge/Car/ZNP(10%) /TEO (10%)	0.504 ± 0.01	16.93 ± 0.4	65.74 ± 1.4	
SR Carrageenan	SRC	0.095 ± 0.001	19.72	32.06	[19]
	SRC/TiO2 (1%)	0.100 ± 0.007	26.58	17.20	
	SRC/TiO2 (3%)	0.105 ± 0.005	20.35	15.00	
	SRC/TiO2 (5%)	0.104 ± 0.003	20.45	19.21	
	SRC/TiO2 (7%)	0.100 ± 0.004	20.86	15.30	
Corn starch	CS/CMC/ZnO (0%)	0.26	-	-	[20]
	CS/CMC/ZnO (3%)	0.28			
	CS/CMC/ZnO (5%)	0.28			
Carrageenan/gelatin	Car/Ge	0.078 ± 0.002	16.64 ± 0.9	84.63 ± 2.2	[22]
	Car/Ge/ oregano essential oil (OEO) (2%)	0.103 ± 0.002	10.09 ± 0.3	87.71 ± 0.7	
	Car/Ge/OEO (2%) /ZnO (2%)	0.093 ± 0.003	21.01 ± 0.5	43.61 ± 0.9	
	SA/ Car/Ge/OEO (2%)	0.133 ± 0.002	30.23 ± 1.6	31.85 ± 1.3	
	SA/ Car/Ge/OEO (2%)/ZnO (2%)	0.127 ± 0.002	41.47 ± 0.8	27.43 ± 1.0	
Chitosan/Wheat	Kontrol	66.93 ± 1.2	6.03 ± 0.2	22.37 ± 1.1	[23]
	TiO2 1%	64.37 ± 1.1	8.80 ± 0.8	17.39 ± 0.9	
	TiO2 1%/Zattaria flower essential oil ZEO	71.31 ± 1.1	8.14 ± 0.4	13.62 ± 1.1	
	TiO2 2%	66.81 ± 1.2	9.44 ± 0.4	12.88 ± 1.3	
	TiO2 2%/ZEO	73.50 ± 1.2	8.71 ± 0.5	11.28 ± 1.7	

Table 2. Barrier Properties in Bionanocomposite Films

Polymer Matrix	Nanocomposites and Essential Oils	WVP (g/m2s Pa)	Solubility (%)	Water Contact Angle (◦)	References
Gelatin/Kappa Carrageenan	Kontrol	8.90 ± 0.31	-	-	[29]
	Gel/KC/SiO2 1%	7.10 ± 0.33			
	Gel/KC/SiO2 3%	2.62 ± 0.33			
	Gel/KC/SiO2 5%	1.61 ± 0.13			
SRkC/Gelatin	SRkC	2,590 ± 0,05	88.50 ± 2.8	82,03 ± 3,2	[30]
	SRkC/SiO2-ZnO	2,424 ± 0,05	78.46 ± 2.4	82,58 ± 7,1	
	Gel	1,625 ± 0,08	65.75 ± 6.0	63,82 ± 10,4	
	Gel/SiO2-ZnO	1,343 ± 0,26	61.30 ± 4.8	74,75 ± 7,3	
	SRkC/Gel 1;3/SiO2-ZnO	2,348 ± 0,01	37.86 ± 4.5	95,80 ± 6,3	
	SRkC/Gel 1;1/SiO2-ZnO	2,395 ± 0,13	43.79 ± 7.2	97,72 ± 4,8	
	SRkC/Gel 3;1/SiO2-ZnO	2,665 ± 0,18	54.58 ± 3.7	83,72 ± 7,3	
Kappa Carrageenan	Kontrol	2.383 ± 0.04	26.32 ± 1.03	-	[31]
	ZEO 1	0.692 ± 0.10	25.88 ± 0.75		
	ZEO 2	0.577 ± 0.09	22.06 ± 0.49		
	ZEO 3	0.425 ± 0.07	17.98 ± 1.66		
	MEO 1	0.945 ± 0.08	23.46 ± 1.23		
	MEO 2	0.665 ± 0.05	20.87 ± 0.94		
	MEO 3	0.496 ± 0.03	16.04 ± 1.35		
Polylactic acid	PLA	103.86 ± 0.22	-	69.72 ± 1.34	[33]
	PLA/fenugreek essential oil (FEO)-5%	104.67 ± 0.58		76.13 ± 1.30	
	PLA/Cur-1%	103.42 ± 0.12		73.12 ± 0.45	
	PLA/FEO (5%)/Cur (1%)	103.00 ± 0.02		80.47 ± 0.61	
Kappa Carrageenan	Kontrol	10.6	95.00 ± 3.79	-	[28]
	KC/ZnO (0,5%)	3.14	82.15 ± 4.63		
	KC/CuO (0,5%)	8.83	71.44 ± 11.18		
	KC/SiO2 (0,5%)	7.51	72.74 ± 11.64		
SR Kappa Carrageenan	SRkC/ZnO (0,5%)	-	61.38 ± 2.47	-	[13]
	SRkC/ZnO (1,0%)		91.85 ± 2.36		
	SRkC/ZnO (1,5%)		83.80 ± 0.65		
			75.04 ± 1.59		
Alginate/Kappa Carrageenan	SA/KC/Clove essential oil (CEO)-(1,5%)	-	59.04 ± 3.06	-	[34]
	SA/KC/ CEO (2,0%)		57.00 ± 1.02		

Polymer Matrix	Nanocomposites and Essential Oils	WVP (g/m2s Pa)	Solubility (%)	Water Contact Angle (◦)	References
	SA/KC/ CEO (2,5%)		54.98 ± 0.99		
	SA/KC/ CEO (3,0%)		52.90 ± 1.47		
Cassava starch	CS	-	-	45.95 ± 0.0	[35]
	CS/Palm Oil (2,5%)			46.83 ± 0.5	
	CS/Palm Oil (5,0%)			61.98 ± 0.2	
	CS/Palm Oil (7,5%)			49.46 ± 0.6	
PBAT (poly (butylene adipate-co-terephthalate)/ Poly (lactide)	PLA/PBAT	-	-	61.61 ± 2.82	[36]
	PLA/PBAT-Clove (1%)			62.24 ± 5.75	
	PLA/PBAT-Clove (5%)			64.69 ± 2.15	
	PLA/PBAT-Clove (10%)			74.74 ± 4.11	

nanocrystals (CNCs) into carrageenan films improved the UV barrier properties of the films, with films having the highest CNC content showing the lowest transmittance due to the UV-blocking properties of the CNCs.

The addition of essential oils (EOs) to bionanocomposites can significantly alter their optical properties. This modification is particularly important for applications in food packaging. As in the study of [39] which states the addition of orange essential oil (OEO) and trehalose significantly affects the transparency of carrageenan-based films. This suggests a synergistic essential oil effect in reducing transparency. In addition, combining OEO and trehalose showed effective barrier properties against UV radiation. This was also the case in the study of [40] which incorporated grapefruit essential oil (GFO) into kappa carrageenan films resulting in a slight decrease in transparency. This was due to the interactions within the polymer matrix and the higher concentration of essential oil, which caused a slight change in the color and transparency of the film.

Transparency values decrease with the addition of essential oils due to the formation of lipid droplets, which scatter light and reduce clarity. This is evident in various studies where essential oils such as oregano and cinnamon have been used in different bioplastic matrices [31,41]. Essential oils contain compounds such as terpenes, which inherently have UV light-blocking properties. Integration of essential oils into the bioplastic film enhances the UV barrier ability, making it suitable for food packaging applications that require UV protection [31,42].

In yam starch-based bioplastics incorporated with clove oil, it was shown that transparency decreased as the concentration of clove oil increased. Higher concentrations of clove oil led to increased interactions between the polymer chains, modifying the refractive index and limiting the passage of light through the bioplastic [43]. Thus, essential oils and nanoparticles also play an important role in enhancing the protective function against UV radiation.

5. EFFECTS OF EO AND NANOPARTICLES ON ANTIMICROBIAL PROPERTIES OF BIONANOCOMPOSITE FILMS

Antimicrobial packaging can reduce and increase the shelf life of food products [44,45]. Packaging with antimicrobial properties is a highly effective technique to inhibit the growth of pathogenic microorganisms in food products, for example, by adding nanoparticles [46]. In some of the literature studies found it was observed that the trend of using silver or zinc nanoparticles is increasing compared to other types of metal nanoparticles on the antibacterial properties of bionanocomposite films. Table 4 shows some of the recent studies related to the effects of adding nanoparticles and essential oils on the antimicrobial properties of bionanocomposite films

Proven in the study [47] in the use of κ-carrageenan/cassava starch-based films to coat fresh strawberries, the addition of ZnO nanoparticles significantly inhibited microbial spoilage and extended the shelf life to 11 days. This is due to the increased surface area of the nanoparticles by contact with oxygen-reactive species that damage the bacterial cell membrane resulting in membrane rupture and preventing bacterial [48]. Similar results were obtained in the addition of Zein nanoparticles to kappa carrageenan/gelatin-based bionanocomposite films showing significant antibacterial effects on E. coli (gram-negative) and S. aureus (gram-positive) bacteria [49]. The antimicrobial activity of the film with nanoparticle incorporation was higher against gram-positive bacteria compared to gram-negative bacteria (E. coli). This is because the cell wall of gram-negative bacteria is more complex and difficult to penetrate, while the cell membrane of gram-positive bacteria is porous and easily penetrated [46].

The integration of essential oils into bioplastic films has been studied for its potential to enhance antibacterial and antioxidant properties. Oregano Essential Oil (OEO)-loaded films showed significant antibacterial activity against pathogenic bacteria commonly present in food including Bacillus subtilis, Escherichia coli, and Staphylococcus aureus. This is influenced by the hydrophobicity of essential oils which can disrupt bacterial cell membranes by interacting with lipids, increasing permeability and causing leakage of vital components, ultimately leading to cell death [50]. In addition, research [45] showed that combining nanoparticles with essential oils effectively increased the antibacterial effectiveness of the film against Escherichia coli and Staphylococcus aureus with the addition of titanium dioxide (TiO_2) nanoparticles and neem essential oil (NEO). In addition, research [51] CA nanoclay film / CEO clove essential oil showed

Table 3. Optical properties in Bionanocomposite Films

Polymer Matrix	Nanocomposites and Essential Oils	UV-Light absorbance (%)	Transparency (%)	Reference
SR Kappa Carrageenan/Cassava starch	SRκC	25.14	62.69	[12]
	SRκC−NPs	6.34	53.33	
	CS:SRκC 1:3	8.05	58.26	
	CS:SRκC 1:1	9.17	61.86	
	CS:SRκC 3:1	9.72	63.57	
Chitosan	Chit/ /chito-oligosaccharide (COS) (1,5%)	-	0.42±0.27	[44]
	Chit/ /chito-oligosaccharide (COS) (2,0%)		0.47±0.27	
PVA/gelatin	PVA/gelatin	14.3	0.48	[37]
	PVA/gelatin/SiO2 (0,5%)	12	0.41	
	PVA/gelatin/SiO2 (1,0%)	15.2	0.35	
	PVA/gelatin/SiO2 (1,5%)	13.1	0.39	
Kappa Carrageenan	Kontrol	-	76.28	[38]
	KC/ Cellulose nanocrystals (CNC) (1%)		63.52	
	KC/ CNC (3%)		48.14	
	KC/ CNC (5%)		46.06	
	KC/ CNC (7%)		21.42	
			21.40	
Kappa Carrageenan	(0,5g) KC/orange essential oil-OEO (0,45 ml)/T80	7.07 ± 1.22	42.06 ± 1.15	[39]
	(0,5g) KC/orange essential oil-OEO (0,45 ml)/T20	10.81 ± 0.04	84.31 ± 0.03	
	(0,3g) KC/orange essential oil-OEO (0,45 ml)/T80	28.23 ± 0.13	87.81 ± 0.35	
	(0,3g) KC/orange essential oil-OEO (0,45 ml)/T20	11.77 ± 0.01	82.14 ± 0.04	
Carrageenan	Car/grape fruit essential Oil-CEO (0,1%)	-	76.13 ± 3.14	[40]
	Car/grape fruit essential Oil-CEO (0,2%)		70.59 ± 2.74	
	Car/grape fruit essential Oil-CEO (0,3%)		67.10 ± 1.56	
Chitosan	Clove oil-CO (0,3%)	-	12.28 ± 1.20	[43]
	Clove oil-CO (0,6%)		10.42 ± 1.10	
	Clove oil-CO (0,9%)		8.67 ± 1.30	
	Clove oil-CO (1,2%)		8.58 ± 1.40	
	Clove oil-CO (1,5%)		5.95 ± 2.90	

Table 4. Antimicrobial properties in Bionanocomposite Films

Composition	Nanocomposites and Essential Oils	Type of Bacteria	Method	Antimicrobial activity of film	References
SR Iota Carrageenan-Fish Gelatin (1:3)/NP SiO2-ZnO	SiO2-ZnO	Escherichia coli dan Staphylococcus aureus	Disc diffusion method	Antibacterial activity of bionanocomposite films increased with the addition of nanoparticles. The antimicrobial activity of the film was higher against gram positive bacteria (S. aureus) than gram negative bacteria (E. coli) this was attributed to the cell wall complexity of gram-negative bacteria compared to gram-positive bacteria.	[46]
Kappa Carrageenan/PVA/Starch/NP ZnO	ZnO (1, 1.5, and 2 % wt/wt)	Escherichia coli Salmonella Typhimurium, dan Staphylococcus aureus	Broth dilution method	Showed growth inhibitory against Salmonella potential E.coli, Typhimurium, and S.aureus microorganisms. This antimicrobial activity may be due to the increased surface area of ZnO NPs and contact with oxygen-reactive species on the nanoparticle surface, leading to the rupture of bacterial cell membranes and inhibiting their replication.	[47]
Gelatin/Kappa Carrageenan/ZNPs	Zein nanoparticles (15 mg/ml)	S. aureus dan E. coli	Disc diffusion method	The addition of Zein nanoparticles showed a significant inhibitory effect against S. aureus and E. coli. For S. aureus, the diameter of the inhibition zone was 8.6 ± 1.3 mm and 10.3 ± 1 mm in E. coli, the diameter of the inhibition zone was 4.2 ± 1.2 mm and 6.2 ± 1.3 mm.	[49]
Starch/CMC/ Oregano essential oil	Oregano essential oil Rosemary essential oil	B. subtil E. coli	Disc diffusion method	Oregano essential oil (OEO) had strongest the antibacterial activity	[50]

Composition	Nanocomposites and Essential Oils	Type of Bacteria	Method	Antimicrobial activity of film	References
	Thyme Essential oil	S. aureus P. aeruginosa		compared to other oils. The Diameter of Inhibition Zone (DIZ) values of OEO against B. subtilis, E. coli, S. aureus, and P. aeruginosa were 39.70 ± 1.80 mm, 29.67 ± 0.75 mm, 55.41 ± 7.47 mm, and 8.62 ± 0.42 mm, respectively. Essential oils are more effective Maroufi et al 2021 Shen et al 2023 gram against positive bacteria because they can directly damage their cell membranes, while the outer cell membranes of gram negative bacteria are more hydrophilic blocking interaction.	
Agar / TiO2 / Radish anthocyanins / Neem Essential Oils	Neem essential oil (NEO)	S. aureus dan E. coli	Disc diffusion method	The addition of Neem essential oil significantly improved the antimicrobial ability against S. aureus and E. coli. This was attributed to abundance antimicrobial the of compounds in NEO (sterols, triterpenoids, active derivatives), and ester which can effectively inhibit microbial growth.	[45]
Cellulose acetate / Nanoclay/Clove Essential Oil	Clove Essential Oil (CEO)	Escherichia coli Listeria monocytogenes dan Lactobacillus sake	Contact method	Addition of nanoclay impregnated with clove essential oil inhibited or reduced proliferation for all the bacterial strains studied. This is related to eugenol which is encapsulated in clove essential oil, a phenolic monoterpenoid compound with a reactive hydroxyl (OH) group. Eugenol can form hydrogen Yang et	[51]

Composition	Nanocomposites and Essential Oils	Type of Bacteria	Method	Antimicrobial activity of film	References
				al 2022 cândido et al-2024 bonds with the active sites of bacterial enzymes. This causes disruption or rupture of the bacterial cell membrane.	
Kappa Carrageenan/SiO2	SiO2 (1 wt%) SiO2 (3 wt%) SiO2 (5 wt%)	S. aureus dan E. coli	Disc diffusion method	SiO2 nanoparticle based films have good antimicrobial activity. The mechanism involves the release of antimicrobial agents from the surface of SiO2 nanoparticles and the application of antimicrobial effects on bacteria in suspension. Therefore, the KCG/5 wt% SiO2 nanocomposite film showed excellent antimicrobial activity against both microorganisms.	[52]
Carrageenan/Orange Essential Oil/Trehalose	Orange essential oil (0.45 mL)	S. aureus dan E. coli	Disc diffusion method	The combination of kappa carrageenan film with OEO showed that it was able to inhibit the growth of E. coli.	[39]

Table 5. Antioxidant properties in Bionanocomposite Films

Polymer Matrix	Nanocomposites and Essential Oils	Methods			Reference
		ABTS	DPPH (AA%)	TPC (mg GAEq/g)	
Chitosan/Gelatin	Chit/Gel	65	76	-	[60]
	Chit/Gel/Rosemary Essential oils (REO) 0.2%	68	78		
	Chit/Gel/REO 0.5%	83	80		
	Chit/Gel/REO 1.0%	85	83		
Polyurethane	PU/ZnO/Nutmeg Oils 25 mikro gram mL-1	64.52 ± 0.10	61.32 ± 0.22	-	[55]
	PU/ZnO/NEO 50	68.24 ± 0.56	65.14 ± 0.52		
	PU/ZnO/NEO 100	69.57 ± 0.80	66.89 ± 0.10		
Carrageenan	Kontrol	18.81 ± 1.23	5.00 ± 0.73	-	[40]
	Car/Gly/Grapefruit Essential oil 0.1%	22.21 ± 0.10	9.46 ± 0.25[b]		
	Car/Gly/GEO 0.2%	26.56 ± 0.51	10.12 ± 0.22		
	Car/Gly/GEO .03%	30.73 ± 0.73	11.36 ± 0.46		
Cellulose acetate	Hybrid Nano clay/Clove essential oil 0	-	3 ± 0[d]	2 ± 0[e]	[51]
	Hybrid Nano clay/CEO 7.5		63 ± 2[c]	36 ± 1[d]	
	Hybrid Nano clay/CEO 15.0		82 ± 1[b]	80 ± 1[c]	
	Hybrid Nano clay/CEO 22.5		86 ± 1[a]	94 ± 1[b]	
	Hybrid Nano clay/CEO 30.0		89 ± 1[a]	104 ± [0]	
Carboxymethyl cellulose/					[58]
Refined Kappa	Kontrol	-	1,05±0,15	-	[53]
Carrageenan	RkC/Lemongrass Essential oil 0.1%		2,32±0,14		
	RkC/LEO 0.5%		6,87±0,09		
	RkC/LEO 1%		15,12±0,06		
Starch	Kontrol	-	4	-	[57]
	S+1.5%ZnO+0.5%Ferulla gumamosa boiss EO		24		
	S+1.5%ZnO+1%EO		39		
	S+1.5%ZnO+1.5%EO		48		
Gelatin	Gelatin	7.8 ± 1.9	5.7 ± 1.3	-	[54]
	Gelatin/MNP0.25%	53.4 ± 2.8	19.6 ± 2.3		
	Gelatin/MNP0.5%	69.6 ± 8.7	42.8 ± 3.9		
	Gelatin/MNP1.0%	85.5 ± 2.5	56.5 ± 4.2		

significant antibacterial activity against Escherichia coli, Listeria monocytogenes, and Lactobacillus sakei as the concentration of nanoclay and CEO increased. This is because Nanoclay can increase the surface area while the essential oil dispersion in the polymer matrix contains the main component, Eugenol, which can disrupt the bacterial cell membrane, increase permeability, and cause cell death.

6. EFFECT OF EO AND NANOPARTICLES ON ANTIOXIDANT PROPERTIES OF BIONANOCOMPOSITE FILMS

Antioxidants are substances that inhibit oxidation reactions caused by free radicals. Antioxidant activity test is needed to determine the content of antioxidant activity in the solubility contained in the packaging film [53]. The addition of natural antioxidants to bioplastics can help prevent oxidation and extend the shelf life of food. Oxidation is the main cause of food degradation, which occurs during processing and storage causing rancidity, deterioration, and the formation of toxic [8]. The Table 5 shows some recent studies related to the effects of adding nanoparticles and essential oils on the antioxidant properties of biocomposite films (Tabel 5).

Nanoparticles can not only affect the physical, mechanical and characteristics of bionanocomposite films but also the antioxidant properties of bionanocomposite films. Literature study by [54] showed that the antioxidant activity of gelatin/MNP nanocomposite films increased significantly with the addition of melanin nanoparticles (MNP) due to the strong antioxidant content of melanin.

The addition of essential oils can improve the characteristics and add antioxidant compounds to bionanocomposite films [53]. The high use of EOs in the production of Bionanocomposites becomes a natural preservative that contains various components and often works synergistically against free radicals and microorganisms to prevent food spoilage and oxidation [40]. In research [55] it was shown that the addition of ZnO nanoparticles and nutmeg oil in the bionanocomposite significantly improved the antioxidant properties of the film. The increase in antioxidant properties is because the nutmeg oil used in the bionanocomposite composition contains polyphenols which are the main components responsible for antioxidant properties. Moreover, the presence of polymer matrix and ZnO nanoparticles has significantly enhanced the antioxidant properties due to their compatible biological and chemical properties [56].

Essential oils are known for their phenolic compounds, such as alpha and beta-pinene, which have strong antioxidant properties. The combination of ZnO nanoparticles and Ferula gummosa essential oil creates a synergistic effect, which further enhances the antioxidant capacity of the film. ZnO nanoparticles themselves exhibit some degree of antioxidant activity due to their ability to interact with and neutralize free radicals [57]. In addition [58] the incorporation of TiO2 nanoparticles and dill essential oil DEO into CMC/MG-based bioplastic films markedly improved the antioxidant properties. Bio-nanocomposite films containing TiO2 nanoparticles and DEO effectively extended the shelf life of refrigerated fish fillet samples. The combination reduced the rate of lipid oxidation and microbial growth. Similarly, the combinations of TiO2 nanoparticles and grape seed essential oil (GSEO) in bionanocomposite films significantly enhanced antioxidant activity. It is known that TiO2 nanoparticles themselves have inherent antioxidant properties due to their ability to neutralize reactive oxygen, when combined with GSEO the nanoparticles help create a synergistic effect, which in turn is very effective in delaying the oxidation process [59].

7. DISCUSSION

The addition of nanoparticles and essential oils to kappa carrageenan-based biodegradable packaging materials has shown significant improvements in various mechanical, physical, barrier, optical properties, as well as their antioxidant and antibacterial properties. Several studies have shown that the addition of nanoparticles such as ZnO and SiO_2 to kappa carrageenan films can improve their mechanical properties. These nanoparticles can increase tensile strength and resistance to elongation, making the film stronger and more flexible. As in the research by [13] found that the addition of ZnO to kappa carrageenan films increased tensile strength but decreased elongation at optimal concentrations. In addition, essential oils such as clove oil and oregano oil can increase film elasticity by interacting with polymer chains [61].

Another important thing that needs to be considered in biodegrdable films is the film's resistance to moisture and oxygen. Nanoparticles function as a physical barrier in the polymer matrix, reducing water vapor and gas permeability by filling gaps in the matrix. This is important for improving the resistance properties of the film. The addition of essential oils also reduces water vapor permeability, providing an additional barrier effect by modifying the film microstructure and reducing water vapor diffusion pathways. Research by [33] showed that the combination of nanoparticles and essential oils in carrageenan films significantly improved the barrier properties against water vapor and gas.

The addition of nanoparticles and essential oils also affected the optical properties of the packaging film. Nanoparticles enhance the UV filtering ability, reducing the transmission of UV light that can damage food products. In contrast, essential oils tend to reduce film transparency due to the formation of lipid droplets that scatter light. As in the study by [39] showed that the addition of orange oil and trehalose in carrageenan films reduced transparency and increased UV barrier properties, which is very useful for protecting food from UV-induced oxidation.

Essential oils are known to have phenolic compounds with strong antioxidant capacity. The combination of nanoparticles and essential oils showed a synergistic effect in enhancing the antioxidant activity of the film. Research by [57] showed that the combination of ZnO and essential oil from Ferula gummosa increased the antioxidant activity significantly compared to the use of one type of additive alone. This is due to the interaction between the nanoparticles and phenolic compounds in the essential oil, which strengthens the antioxidant capacity of the film. Studies combining nanoparticles and essential oils showed a synergistic effect in enhancing the antioxidant properties of the films.

Bionanocomposite films with nanoparticles and essential oil also showed better antibacterial properties. ZnO nanoparticles have significant antibacterial activity against gram-positive and gram-negative bacteria. In addition to nanoparticles, essential oils with compounds such as eugenol and carvacrol are also effective in inhibiting bacterial growth by disrupting bacterial cell membranes. Research by [45] showed that the combination of TiO_2 and neem essential oil increased the antibacterial activity of the film against Staphylococcus aureus and Escherichia coli.

Overall, the addition of nanoparticles and essential oils to carrageenan-based biodegradable packaging films provided significant improvements in their mechanical, physical, barrier, optical properties, as well as their antioxidant and antibacterial properties. This combination results in a packaging material that is not only environmentally friendly but also provides better protection for food products, making it a potential solution for future food packaging applications.

8. CONCLUSION

The preview of this review demonstrates the addition of nanoparticles and essential oils significantly enhanced the properties of kappa-carrageenan based bionanocomposite films. The mechanical, optical, barrier, antibacterial, and antioxidant active characteristics of kappa-carrageenan based bionanocomposite films were significantly increased by the addition of nanoparticles and essential oils. By establishing a more complex matrix, nanoparticles increased the films' tensile strength and moisture resistance, while essential oils added to bionanocomposite films improved their flexibility, antibacterial and antioxidant properties. Strong synergy between essential oils and nanoparticles produces bionanocomposite films with enhanced barrier capabilities and balanced mechanical properties, which may find use in smart and ecologically friendly food packaging.

DISCLAIMER (ARTIFICIAL INTELLIGENCE)

Author(s) hereby declare that NO generative AI technologies such as Large Language Models (ChatGPT, COPILOT, etc) and text-to-image generators have been used during writing or editing of manuscripts.

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COMPETING INTERESTS

Authors have declared that no competing interests exist.

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