Effects of incorporation of essential oil into film for fruit packaging application

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Abstract. Biodegradable composite films with antimicrobial properties consists of three biopolymers (polysaccharides, proteins, fats), additives (plasticizers, emulsifiers), and synthetic or natural antimicrobial. Biodegradable packaging embedded in food products with antimicrobial agent such as essential oils (EOs) is an innovative green technology based on a novel approach to substitute conventional and synthetic petroleum packaging. Good packaging is necessary to prevent spoilage during postharvest stage. Introducing EOs onto the fruit packaging films strongly inhibiting bacterial and fungal activities on the fruit products and gives positive effect on the storage of the products. This review focuses on the contribution of the incorporation of EOs and biodegradable films to prolonging the shelf life of fruit products in addition to reduce the risk of growth of pathogenic bacteria on the surface of fruit products. Essential oils incorporated into film provides a new strategy and improves efficiency of fruit packaging film in enhancing quality and shelf life of post-harvest fruit products.

1. Introduction

In recent years, recycled plastic films is a disposable material for the food packaging industry. This raises environmental issues as most plastics are naturally not biodegradable and the amount of synthetic plastic waste generated by the food packaging industry increasing over the years [1]. Therefore, the food packaging industry are encouraged to develop a new biodegradable packaging material. In the strive for alternative materials to plastics, scientists have placed particular focus on biopolymers. One of the most critical aspects, accessibility, is the fulfilment of certain abundant natural polymers, such as cellulose (and derivatives like carboxymethyl cellulose (CMC), methyl cellulose (MC), cellulose acetate (CA), etc.), chitosan, lignin, starch, polylactic acid (PLA), polyvinyl alcohol (PVA), alginic acid and derivatives etc. In producing biodegradable polymer films, the addition of active compounds, such as essential oils (EOs), may enhance their functional properties, (e.g. mechanical properties and antimicrobial properties), which prolong the shelf-lives of packaged food. Moreover, incorporating EOs,
which is made from the extraction of plants and spices, into biodegradable film is able to improve its applications in food packaging industry. For food packaging industry, EOs exhibits antimicrobial and antioxidant properties and is classified as Generally Recognized as Safe (GRAS) [2], [3]. Due to its strong flavour, the application of EOs as food preservatives is limited, and therefore can be used for food packaging by integrating EOs into edible films. In this review, an evaluation is carried out by evaluating the effect of incorporating EOs onto the biodegradable film on the films’ structural effect, antioxidant capacity, antimicrobial capacity, gas transfer behaviour, and its real application in the food packaging.

2. Structural effect
The structural arrangement of the components in the film-forming dispersion can influence the microstructure of the film [4]. Sánchez-González et al. (2009) formulated films with different antioxidant agents, which include EOs. EOs causes discontinuities (EO droplets) and hence causes the films to have a wider and thicker structure, in comparison to films without the addition of EOs [5]. It is commonly observed that the final microstructure of the film is determined by the structural arrangement of the components in the film forming dispersion. Furthermore, their development during the drying process plays an important role, given that unstable phenomena such as droplet flocculation, coalescence and cremation can occur. However, not only the essence of the EO, but also the homogenization technique determines the mechanisms that affect microstructural changes in the film. In this framework, important structural variations have been identified as the size of the EO droplet has been reduced [6]. Apparently, the droplet reduction favoured the intimate incorporation of the EO into the polymer matrix, resulting in increased interactions between the polymer and the oils, a less stable polymer matrix and barely visible oil droplets. Furthermore, previous studies reported a plasticizing effect occurs when phenolic compounds in the EOs is incorporated into the polymer matrix [7]. Moreover, some previous studies show that the tensile strength (TS) of the film increases was due to the incorporation of EOs [8]. A possible explanation of the strengthening effect is linked to the ability of EO to induce the rearrangement of the polymer network. The films incorporated with Zataria EO had relatively smoother surfaces compared to the control starch films. However, several holes appearing to be uniform in size have been found in these films and this is possibly due to the presence of essential oils in the film formulation which may result from the volatility of the essential oil compounds [9]

3. Antioxidant
As oxidation lower the quality of the packaged fruit, the incorporation of natural antioxidants into packaging materials has become popular. Applying natural antioxidants onto the film of the packaging material to improve the packaged fruits’ shelf-life has become more preferable by the consumers as synthetic antioxidants potentially causing detrimental effects on their health. As antioxidant agents, EOs have different modes of action. As an antioxidant agent, phenolic compounds in the films scavenging active oxygen and nitrogen species at the surrounding of the packaged fruits, hence prolong its shelf-lives. In a DPPH test conducted by Siripatrawan et al. (2010), the antioxidant capacity of the chitosan films increases when green tea extract is incorporated on the surface of pure chitosan film [10]. Besides, it is stated that the interactions between biodegradable films and polyphenolic compounds in EOs play a vital role in the modifying of the film properties in enhancing antioxidant capacity. However, the quality of EOs as an antioxidant can decline in the presence of oxygen. A study conducted by Bonella et al. (2013) stated that polyphenol compound in EOs oxidizes to form polyphenolic oxidase, which causes enzymatic browning in the packaged fruits. Therefore, to prevent the enzymatic browning, the films and coatings in the packaging material is required to dark-coloured pigments on the packaged food products in the presence of oxygen [11]. Polylactic Acid films containing different type of essential oils (thyme, rosemary, oregano) might serve as effective antioxidant packaging materials for the preservation of food [12]. PLA/PTMC/OEO composite films containing 9 wt % exhibited strong antioxidant effects [13]
4. Antimicrobial
Antimicrobial packaging has into two group: (1) biodegradable packaging and (2) non-biodegradable packaging. Biodegradable packaging includes edible coating and films made of proteins, lipids and starch, chitosan, polyactic acid, polyhydroxy butyrate, and polyhydroxyalkanoate. The level of antimicrobial activity in the films is determined by observing the ability of EOs to inhibit microorganism growth, in which by having the bioactive compounds of EOs interacting and, altering the permeability of cell membranes, which causes the leakage of cell content, which contain vital components for the survival of microorganisms [7]. Numerous studies have assessed that antimicrobial packaging can inhibit the targeted bacteria effectively when a sufficient amount of antimicrobial agents are integrated into packaging materials [14]. According to the result in the previous study, the films without the addition of EOs showed no activity against tested bacteria, but the film that blended with essential oil inhibited the bacterial activities in the films [9]. Qin et al (2016) have use bergamot essential oil in their studies and observe that the films contain essential oil can maintain the quality of mango and extend the shelf life to 15 days [7]. In Priyadarsh et al (2018) studies, they added the Apricot kernel essential oil to the chitosan films, thus the results showed that the films with essential oil successfully inhibited the fungal growth on packaged bread slices as compared to one without essential oil [15]. The authors have blend the Polylactic acid films with Origanum vulgare L. Virens essential oil in their studies, they observed the films exhibited in vitro antibacterial activity against Staphylococcus aureus, Yersinia enterocolitica, Listeria monocytogenes, Enterococcus faecalis and Staphylococcus carnosus [16]. Table 1 are presented some types of antimicrobial packaging materials, their main polymer, antimicrobial agent, and the observations.

Table 1. Antimicrobial packages and their applicability.

| No. | Type of films   | Type of essential oil                        | Observations                                                                 | Ref  |
|-----|----------------|---------------------------------------------|------------------------------------------------------------------------------|------|
| 1.  | Polylactic Acid| Bergamot EO                                 | Maintain the quality of mango and extend the shelf life to 15 days           | [7]  |
| 2.  | Starch         | Zataria multiflora Boiss (ZEO),             | Film that blended with essential oil inhibited the bacterial activities in the films | [9]  |
| 3.  | Chitosan       | Apricot (Prunus armeniaca) kernel EO        | Successfully inhibited the fungal growth on packaged bread slices.            | [15] |
| 4.  | Polylactic Acid| Origanum vulgare L. virens EO               | Films exhibited in vitro antibacterial activity against Staphylococcus aureus, Yersinia enterocolitica, Listeria monocytogenes, Enterococcus faecalis and Staphylococcus carnosus. | [16] |

5. Gas transfer
Another important characteristic that influences quality of the packaged products is the ability of films to retard moisture loss from the packaged product. Water vapour and oxygen barrier properties of packaging are critical in the food industry, same goes to the permeability of CO2 and aromatic compounds also plays an important role. The water vapour transfer processes in films depend on the hydrophilic-hydrophobic ratio of the film constituents [17]. Thus, the incorporation of hydrophobic EO into hydrophilic polymer matrices might result in the improvement of their water vapour barrier properties. This trend was observed by Sánchez-González et al. (2009) in HPMC films with tea tree EO [5], by Zeid et al. (2019) in Polylactic acid films with different EOs (thyme was the most effective)[12]
and also by Nur et al (2017) who incorporated cinnamon EOs into Polylactic acid biocomposite films [18]. However, as complex mixtures of various chemical compounds, the hydrophobicity of EOs is a variable feature, affecting their effectiveness as water vapour permeability depressors. Chi et al. (2019) developed Polylactic acid films with bergamot EOs and noted that they had a different impact on water permeability, which was due to their varying properties [19]. Some works reported that the incorporation of higher amounts of plant EOs increases the oxygen permeability values [20]. Low levels amount of EOs shows no significant differences. A possible reason is that low levels amount of EO is not sufficient to induce the changes in the film structure that promotes the increased mobility of oxygen within the film structure itself [9]. In contrast, Han et al. (2006) reported that the pea-starch film structure changes as the beeswax concentration increased to exceed 40%. The author suggested that when a higher concentration of EO is added to the film, the oxygen molecules possibly penetrating through the film between the EO particles, which in turn providing oxygen-penetration channels [21]. Besides, the extent of the barrier is influenced by the chemical properties of the material [22].

6. Real application of EOs into films for food packaging
Many polymers are widely applied in food packaging. These polymers can be incorporated with synthetic or natural additives which are responsible for showing their function [23]. In the application of food packaging, Priyadarshi et al (2018) attempt to evaluate the effect of kernels of apricot (prunus Armeniaca) EOs concentration on the chitosan films’ physicochemical and functional properties. They also compare the physicochemical and functional properties of EOs-incorporated chitosan films with that properties of neat chitosan films. The results shows that the EOs-incorporated chitosan films has an excellent antimicrobial and antioxidant properties in comparison to the antimicrobial and antioxidant properties of the neat chitosan films [15]. Next, Sánchez-González et al. (2011) studied the antimicrobial properties of chitosan film with various concentrations of bergamot, lemon and tea tree EOs [24]. Inoculated agar plates were used as model food systems and periodic microbial counts showed the antimicrobial activity of the films. Sánchez-González et al. (2011) has proven the antimicrobial activity of HPMC or chitosan films incorporated with bergamot EO applied to grapes. EOs may have both qualitative and quantitative variations due to a wide variety of bioactive compounds resulting in variable biological efficacy. Moreover, the oil is not only able to retard microbial growth, it can also maintain sensory acceptability when applied to packaging fresh fruits and vegetables [25].

7. Conclusion
In conclusion, incorporating essential oil on the film affects the continuity of the polymer matrix and hence changing the physical changes of the polymer matrix, depending on the level of interaction between the polymer-oil components. Generally, adding essential oil to the film can weaken the film structure, strengthening the gas barrier, and reducing the films’ transparency. Furthermore, the addition of essential oils in the film can provide the film antioxidant and antimicrobial properties that can keep the packaged-product quality. The antioxidant activity depending not only on the specific antioxidant activity of the oil compounds, but also with the permeability of the film to oxygen. Incorporation into biodegradable films can promote the antimicrobial capacity of EOs, and the efficacy of the biodegradable film against microbial growth will depend on the nature of the oil and the form of microorganism. The composition of EOs and its specific interactions with the polymer can determine its effectiveness as an active ingredient. Thus, showing the potential of biodegradable polymeric films embedded with the essential oil to substitute the conventional packaging in reducing environmental hazards and prolong the packaged fruits’ shelf-life. However, more work is required in this area of research in order to maximise the effectiveness of the bioactive incorporated agents.

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**Acknowledgements**
The authors would like to thank the Ministry of Education for financing the research under Fundamental Research Grant Scheme FRGS/1/2018/STG01/UNIMAP/03/ and Faculty of Chemical Engineering Technology, Universiti Malaysia Perlis (UniMAP).