



Review article

Potentials of Berry Fruits Pomaces for Bio-Based Films

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Abstract

Nowadays, there are new approaches and goals for packaging materials for advantages on foods and friendly properties on environment. For these purposes, biodegradable polymers are much preferred to replace conventional polymeric goods in packaging applications. Biopolymers, such as carboxyl methyl cellulose (CMS), environmentally friendly sustainable plastic alternatives. The worldwide need for bioplastic as an alternative for conventional plastics because of their nontoxicity, biocompatibility, renewability, and biodegradability properties. They are commonly produced using different raw materials (proteins and polysaccharides), which are mostly derived from plants (cellulose-based plastics and starch derived plastics), and microbial sources.

Moreover, there is more than a need to preserve natural resources and to reduce plastic waste materials by replacing them by bio-alternatives. It should be solution that wasted foods, which are the GRAS, should be a source for enriching biopolymers with their valuable bioactive compounds. Fruits and vegetables, especially berries that contain high content of valuable bioactive compounds, can be used for enriching to biopolymers and for production of active films and indicators. It has high importance for food science because oxidation and microbial contamination present major problems that influence quality and safety of different products during their storage. Currently, to overcome these points, there are some approaches can be applied. For instance, bioactive compounds and antioxidant agents from berries can be directly added to biopolymers. Blueberries, red and purple grape, black mulberries etc. and their industrial wasted present good sources of antioxidants, non-flavonoids and flavonoids. Phenols are primarily found in the skin and pomace of berries. It is known that most of these phenols are typically wasted due to poor extraction during fermentation. Therefore, these wastes are of great importance for biofilms. However, it is also essential to investigate their effects on other properties, such as texture, taste, solubility, etc.

Keywords: Antioxidants, Berry Fruits Pomaces, Carboxyl Methyl Cellulose (CMS), Enriched Biopolymers, Packaging Materials, Sustainable Plastic Alternatives.

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INTRODUCTION

The plastic materials are very critical due to its potential character to people and whole ecosystem. Therefore, an uncontrolled increase in population with excessive non-renewable resource exploitation is resulted in cumulative waste volume¹⁻³. Plastic materials are mostly used as packaging in the food industry. Alternatively, the bio packs assumed to have similar properties with traditional plastic packages. Besides, it is accepted that biodegradable plastic holds excellent characteristic properties and is more reliable nowadays³. The developed cellulose-based bioplastic materials are one of the best possible solutions for the new approach that includes the use of traditional plastics and is called zero waste around the globe⁴. Biodegradable plastics derived from renewable biomass resources such as agricultural by-products, vegetable waste, fruit waste, biopolymers, and microorganisms. Biodegradable plastics can be broken down into a smaller part through biological treatment either by applying aerobic or anaerobic techniques, and the process is generally defined based on the types of bioplastics such as starch, cellulose, biopolymers-based materials^{3,5}.

It is known that there are different requirements of food packaging materials such as avoiding possible contamination etc. Also, the properties of the package materials depend on the mechanical strength, permeability to oil, water, oxygen, microbial action, etc. during storage and their distribution. Likewise, other essential factors are concerning recyclability, material costs, disposable nature, and sustainability⁶.

In upcoming years these biodegradable polymers are much anticipated to replace conventional polymeric goods in packaging applications⁷⁻⁸. There are lots of research about biodegradable packages. However, it has also great importance to characterize the acceptance of aroma and flavours compounds and antioxidant properties that limited work has carried out^{9,10}. This is mostly because of the oxidation and microbial contamination present major problems that influence quality and safety of different products during their storage. Oxidation can cause the loss of sensorial and nutritive quality of packed food, and in negative conditions it can be resulted with formation of toxic aldehydes¹¹⁻¹². The skin and pomace of berries, such as blueberry, raspberry, black and purple mulberry, red and purple grape etc., are good sources for antioxidants. Besides, it has been proven that high percent of antioxidant of berry left in pomace after pressing¹³. Furthermore, berries are considered as the world largest crops and rich sources of non-flavonoids (hydroxybenzoic acid and its derivate) and flavonoids (anthocyanins, flavan-3-ols monomers and polymers, flavonols and dihydroflavonols)¹¹. Even though fruit pomace is rich source of antioxidants, the main drawback is their low stability. Recovery of natural bio-actives from berries is generally obtained with solid liquid extraction, heating or grinding. However, these techniques have some disadvantages to the extraction performance and phenol stability as well as the harmful effect environment¹⁴. For this purposes, green extraction techniques can be helped.

The design of the bio-films composition which enriched with berry pomace is important because to obtain richer biofilms¹⁵. Different hydrocolloids have good film-forming properties, and they are often studied as bio-sourced material suitable for different packaging applications, some of them being edible¹⁶. Recently, the literatures about combination of carboxymethyl cellulose (CMC) and chitosan (CS) with skin pomace extracts of berries prepared by novel extraction techniques is not enough. CMC is one of the most important cellulose ethers used in food and some other industries¹¹.

Agro-Wastes and Potential Usage Areas

In the food industry, especially in the fruit and vegetable processing sector, significant amounts of biowastes such as peels, pomace and seeds are generated as by-products. These wastes contain high levels of macro components such as nitrogen, phosphorus and carbon, as well as a considerable amount of water activity¹⁷.

They contribute to both solid waste accumulation and fermentation, leading to environmental pollution both physically and microbiologically. Despite this, biowastes are rich in substances like pectin, starch, cellulose, agar, gelatin, alginates, carrageenans, flavonoids and antioxidants. Due to their richness in these compounds, biowastes can be transformed into high-value products by utilizing them as raw materials in various fields, without compromising their original meaning¹⁸. Approximately 50% of oranges used in the orange juice processing industry are discarded as by-products such as peels and pulp. These by-products constitute about 95% of the biomass classified as waste and contain water, organic compounds and valuable substances. According to the information available in the literature, these biomass materials consist of various components, including pectin, soluble sugars, cellulose, starch, proteins, lignin, ash, fats and aromatic compounds. The orange peel is considered a potential waste source for biopolymer production due to its high content of starch, pectin and fibers¹⁹.

Yaradoddi et al. (2022) investigated to the production of biodegradable packaging material from orange waste. In this study, the high pectin and cellulose content of orange peels formed a durable film; additionally, the oil content and added glycerol increased the adhesive strength of the film. It was observed that the obtained film had a hard and brittle structure²⁰.

Therefore, flexibility needs to be addressed for its use in commercial applications and sustainable packaging. In order to the produced film to be used as a packaging material, its hydrophobicity and liquid retention capacity are also very important for softening the product. It was stated that the films obtained from orange peel decompose 90% anaerobically in approximately 15 days, so they can be used as a short-lived packaging material¹⁹. However, characterization and requirements must be made in order to achieve the desired level of other properties of packaging, such as hygroscopic properties.

Another fruit has usability for packaging material is banana that contains 60% fruit and 40% peel. Banana peel is rich in organic products such as cellulose (7.6-9.6%), hemicellulose (6-9.4%), pectin (10-21%) and lignin (6-12%). The reproducibility of banana peel is high at 18.5%, so it can be seen as a content from which film can be produced. Studies have been conducted on the production of cellulose-based films from waste banana peels and stems, which have high cellulose content, to evaluate them as biodegradable packaging materials. Ai et al. (2021) examined to the use of cellulose films synthesized from banana stem fibers in preserving mangoes, it was observed that the enhanced permeability of cellulose films could delay post-harvest ripening in climacteric fruits, prolonging storage time. The study found that the residue rate of the cellulose film left in the soil after a four-week period was 7%, indicating the high biodegradability of the cellulose film. Consequently, it was concluded that the cellulose film could serve as a potential packaging material to keep climacteric fruits such as mangoes fresh, contributing to extending their shelf life²¹.

Pomegranate processing industry generates approximately 55% pomegranate peel as a by-product. Pomegranate peel is rich in pectin, tannins, antioxidants, and antimicrobial compounds. In a study where pectin extracted from pomegranate peels was used as the film matrix, different concentrations of montmorillonite clay were added to the formulation, and the mechanical and barrier properties of the films were examined. An increase in montmorillonite clay concentration led to a 40% reduction in water vapor permeability and a decrease in elongation at break during tensile testing²².

Pirsa et al. (2020) aimed at monitoring the shelf life of cream cheeses, an antibacterial and biodegradable film was developed using a material composed of chitosan, pomegranate peel extract, and *Melissa officinalis* extracts. This film incorporates the antimicrobial and antioxidant properties of pomegranate and the antibacterial and antifungal effects of *Melissa officinalis*. Additionally, chitosan's antimicrobial activity was included in the formulation. The resulting biodegradable film was specifically designed for monitoring the spoilage of rapidly perishable products like cheese. Due to the presence of anthocyanin pigments in the film, the sensitivity of pomegranate peel extract to pH changes enables the control of pH variations²³.

In another study, citrus peel and potato pulp, obtained from food industry waste, were mixed in different ratios to develop a biodegradable film. The proportions of components used in the film formulation are crucial factors that significantly influence the film properties. Potato pulp is a rich source of starch, cellulose, hemicellulose, and fermentable sugars, making it suitable for biopolymer film production. Lemons are generally consumed fresh or processed into juice, with the peel being particularly rich in flavonoids, pectin, and essential oils. Moreover, lemon peels have been noted in the literature for their antioxidant and antimicrobial properties²⁴. In this research, the effect of particle size of the waste materials on the film properties was investigated, in addition to the evaluation of the waste utilization. For this purpose, ultrasonication was applied to film solutions to reduce particle size and

prevent aggregation of waste particles. A film made entirely of lemon peel showed a 100% reduction in water vapor permeability compared to a film made entirely of potato pulp, indicating a 73% decrease. The addition of lemon peel significantly reduced the water vapor permeation of the films. Films derived from potato pulp and lemon peels exhibited the lowest water vapor permeability and acceptable mechanical properties when the potato-to-lemon ratio was 0.5:1. Changes in particle distribution, size, and shape were reflected in the mechanical properties of the films. As the amount of lemon peel in the formulation increased, the elongation at break also increased. Furthermore, decreasing potato pulp and increasing lemon peel powder led to a reduction in reddish and yellowish hues in the films²⁵. The film developed in this study with a potato-to-lemon ratio of 0.5:1 was used for bread packaging. Slices of bread packaged with the biodegradable film were compared with those without packaging and those packaged in polyethylene over a 5-day shelf life. At the end of the 5-day period, bread slices wrapped in the biodegradable film experienced more weight loss compared to slices in polyethylene packaging but less than the unpackaged slices²⁵.

Bio-Based Films: An Eco-Friendly Alternative

Bio-based films are novel products and alternatives to conventional plastic films based on fossil resources, especially for single-use applications like food packaging. Conventional fossil-based plastic films offer advantages in terms of high performance, low costs, easy production, and well-established manufacturing methods; however, bio-based films may excel in terms of carbon footprint, biodegradability, and active properties (such as antioxidants and antibacterial properties)²⁶.

Berry Fruits Pomaces: Composition and Characteristics

Berry pomace compositions for biomaterials

Berries are brightly coloured, juicy, sweet or sour fruits which may contain numerous seeds and they generally are processed into juice results in approximately 20-30% by-product. The pomace from berry juice processing contains high content of dietary fibre and bioactive compounds. Besides, the berry pomace is an attractive source for nutrients because of exposing to minimal pesticide level. The health promoting effects of bioactive compounds, such as cancer prevention, anti-inflammatory activity, reduction of cardiovascular disease, reduces oxidative stress, support on immune disorders etc., is a well-known data anyway^{27, 28}.

Berries have skin, pericarp, intracellular juice, and seeds with different fractions depend on type and cultivar. They have high content of moisture and, after drying process they have significant percent dietary fibre from cell wall components. Several berries are composed of lignin, hemicellulose, cellulose, and pectin. Berries contain high amounts of anthocyanins and polyphenols. Fruits are rich in phenolic acids, stilbenes, and flavonoids, with some of these components, particularly anthocyanins,

known to be responsible for the colour of the fruit and their high antioxidant capacity²⁹. After pressing, skins and seeds remain in the pomace and with them the fractions of antioxidants that are associated with cell wall compounds²⁸.

Berry pomace is usually recycled as animal feed, composted, used for biogas production, or simply discarded. However, the pomace must be evaluated in short periods because berry pomace has a moisture content of approximately 50% and as it contains various digestible nutrients, and it has high risk for microbial spoilage³⁰. For this reason, the most recent research studies dealing with the utilization of berry pomace for bio-based films in food packages. In addition, the researchers obtained data about berries pomaces have enhanced the physical, mechanical, antioxidant, and antimicrobial properties of packaging and bio-composite systems^{28, 31}.

The by-products and pomaces of berries can be valuable sources of bio-polymer materials. The recent literatures have focused on biopolymers, bio-composites, active packaging, and by-products and the innovative technologies used for bioactive compound extraction. The mechanical, thermal, antibacterial, and physicochemical properties of berry-based biomaterials have recently shown improvement. Common matrices used for biomaterials are carboxymethyl cellulose (CMC), polylactic acid (PLA), polybutylene succinate (PBS), polyethylene (PE), polyhydroxyalkanoate (PHAs) etc. Additionally, gelatin, starch and pectin are an extracted compound used in the polymeric matrix, and the films produced are both active and biodegradable³²⁻³³.

Utilization of berry wastes in food packaging systems

The growth of berry-enriched polymer materials has been rapid, and their applications in active packaging, biodegradable film, pH indicator, bio-composites, by-products, and recent technologies for the extraction of bioactive compounds mean they appear to have a promising future in the coming years³¹. Luchese et al. (2018)³⁴ added blueberry pomace into cassava starch films. The aromatic compounds in blueberry pomace improved the light barrier properties, indicating the films' ability to preserve food against UV lights. Besides, the films were structurally stable when immersed in water for more than 24 h. The researchers suggested the feasibility of the films for packaging aqueous food products. In another research, the addition of antimicrobial additives into the packaging materials can reduce the food spoilage and microorganism contamination. They studied with hydroxypropyl high-amylose starch biopolymer and wasted pomegranate peel for *S. aureus* and *Salmonella* bacteria³⁵. De Moraes Crizel et al. (2016)³⁶ were used fiber and ethanolic extracts from blueberry juice from processing waste to make active films from gelatin capsules wastes. They evaluated to the applications for sunflower oil and, they improved light barrier properties; reduced the lipid oxidation of sunflower oil and stability in antioxidant activity. Shukor et al. (2021)³⁷ prepared tapioca-starch-based films incorporating thymol, jackfruit skin, and straw as filler cherry tomato. They obtained an improvement in mechanical and barrier properties was observed for the films. Besides, the treatments inhibited to

bacterial and fungal growth due to the antimicrobial activity of the films. Priyadarshi et al. (2018)³⁸ added apricot kernel essential oil into chitosan and, they evaluated to applications in bread slices. They noted that the apricot kernel essential oil improved water resistance, water vapor barrier, and mechanical properties and, inhibited *E. coli*, *B. subtilis* and fungal growth. Lin et al. (2020)³⁹ enriched to Pectin, sodium carboxymethyl cellulose (CMC–Na) with thyme essential oil and they studied to the effects on Okara soluble dietary fiber. Similarly with other research, they detected that higher mechanical, barrier, optical, and antioxidant properties. However, they also reported that the samples had not significant antimicrobial activity against *E. coli* and *S. aureus*. Several literatures presented the positive effects of berries on biopolymers in different applications.

Applications of Berry Fruits Pomace Bio-Based Films

Scientific research on bio-based materials with antioxidant properties has become one of the primary areas of interest for the scientific community. This interest is largely due to the fact that oxidation and microbial contamination pose significant challenges, particularly affecting the quality and safety of food products during storage. Oxidation can lead to sensory and nutritional quality losses in packaged foods and, under extreme conditions, can result in the formation of toxic aldehydes. Currently, two commercially employed methods are used to address these issues: direct addition of antioxidant agents to food or packaging them in a modified atmosphere. At this point, active antioxidant packaging, which focuses on ensuring controlled release of antioxidants during storage, has gained significant attention^{40,41}. Given the global trend to reduce the use of synthetic additives, natural sources of antioxidants are taking the lead⁴².

Bio-Based Films

Synthetic plastics derived from petrochemical resources have been indispensable elements of modern life since the twentieth century. Additionally, due to their low cost, versatility, lightweight nature, and ease of processing, they are the most commonly used packaging materials. However, the accumulation of non-biodegradable plastics in landfills and water sources is becoming an increasingly serious environmental issue⁴³. Traditional synthetic polymers like polyethylene or polypropylene are being replaced by biopolymers obtained from natural and sustainable sources. Biobased polymers such as polysaccharides, proteins, lipids, and their composites are considered the most promising candidates for reducing environmental impact arising from waste accumulation because these materials are biologically degradable and offer the advantages of being abundant and renewable⁴⁴. Edible and biodegradable films must possess various characteristics to meet specific functional requirements, including moisture barrier, solute and/or gas barrier, water or lipid solubility, colour and appearance, mechanical and rheological properties, non-toxicity, and more. These properties can vary depending on the type of material used, the manufacturing process, and the intended applications. To enhance the functionality of the film, additives such as plasticizers, cross-linking agents, antimicrobials,

antioxidants, texture agents, and similar substances can be incorporated to improve its properties⁴⁴. Films generally need to be resistant to breaking and abrasion (to strengthen and protect the food contents) and flexible (to accommodate possible deformation without breaking)⁴⁶. The mechanical properties of edible films and coatings depend on the type of film-forming material, particularly its structural compatibility. The mechanical properties are linked with the film forming conditions, e.g. type of process and solvent, cooling or evaporation rate, etc., and the coating technique (spraying, spreading, etc.)^{45,47}.

Conclusion

Berry wastes from the food industry result in environmental pollution; however, these wastes can be valuable sources of polymer materials. This review focuses on recent advances in biofilms, biopolymers, and by-products used for bioactive compound extraction. The mechanical, thermal, antibacterial, and physicochemical properties of berry-based biopolymers have recently shown improvement. By-products of berries has the potential to improve polymer properties, but it is not forgetting to sustain the cost and energy benefits while also maintaining the mechanical and thermal properties.

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