

Green Biomaterials: Applications of Plant-Derived Biofilms

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Abstract. The use of plants as biomaterials holds an extremely high potential. The application of plant materials in several areas of agri-food, biotechnology, engineering, and health sciences is of enormous interest to the green and low-carbon industry. Plants are a source of polysaccharides, essential oils, and vitamins, among other components that can be used in the formulation of composite materials of plant origin. Polysaccharides with good mechanical properties include alginate, cellulose, and pectin, which already have been applied for drug delivery, production of artificial extracellular matrices, and 3D scaffolds for tissue engineering. Moreover, the extracts and essential oils of some plant's present anti-inflammatory, antioxidant, and antibacterial capacity. These bioactive properties can be transferred to applications in the most diverse areas, through encapsulation of plant extracts/essential oils into biofilms or polymeric matrices. Plant-derived biomaterials are generally highly available and easy to produce at low costs. Although biodegradable, they decompose at slower rates than other biomaterials due to their enzymatic resistance. In addition, biomaterials of plant origin show high biocompatibility, low immunogenicity, and additional nutritional value. Here, we briefly review on the applications of enriching and encapsulating plant derived extracts into natural polysaccharide-based materials. The use of natural materials is an efficient solution towards higher environmental sustainability and a greener economy.

Keywords: Biomaterials · Plants-derived materials · Polysaccharide-based biofilms · Alginate · Pectin · Cellulose · Encapsulation

1 Introduction

Natural polymers such as polysaccharides are valuable assets due to their biodegradability, biocompatibility, and nontoxicity. Polysaccharides of algae and plant origin such as alginate, cellulose and pectin are outstanding materials that can be accessible at low prices and show tunable physicochemical and mechanical properties, along with high renewability and recyclability. Moreover, functional additives such as antimicrobial compounds, antioxidants, minerals, vitamins, and other bioactive molecules can be added into polysaccharide-based matrices to synergistically extend the favorable properties of these biopolymers. Thus, the functionality of polysaccharide-based materials can be further enhanced through incorporation of other green materials, such as plant extracts, polyphenols, and essential oils, to be used in a wide range of applications (e.g.: smart materials, biosensing, wastewater treatment, bioremediation, food coating, wound dressing, drug delivery and tissue engineering) (Fig. 1).



Fig. 1. Polysaccharide-based enriched biofilms: extraction of compounds, preparation of biofilms and fields of application (e.g., bioremediation, drug delivery, wound dressing, tissue engineering, food coating and 3D scaffolding). Image produced by E. Foulquié with Biorender.com.

1.1 Alginate-Based Enriched Biofilms

Alginates are unbranched polysaccharides extracted from brown marine algae and bacteria [1]. Alginates are copolymers of β -D-mannuronic acid (M) and α -L-guluronic acid (G) linked in an irregular block-wise manner, that differs significantly according to the source of extraction [1]. Alginates can undergo gelation and form transparent water-soluble structures that can be processed into hydrogels, microspheres, foams, fibres, gauze, or sponges [2]. Crude extracts, essential oils, and bioactive molecules of plant origin can be loaded into these alginate-based supports, towards the production of highly versatile eco-friendly structures, with various applications [3–13]. In what regards food coating, alginate-based edible coatings enriched with Ficus hirta fruit extract inhibited the growth of pathogenic fungi in Nanfeng mandarins, stored for 100 days at 6 °C [3]; while alginate coatings enriched in pomegranate peel extract delayed the ripening of guavas (*Allahabad safeda*) during 20 days of storage, at 10 °C [4]. Additionally, other authors have shown that alginate coatings enriched with green tea and grape-seed presented antiviral properties against norovirus and hepatitis A virus [5], whereas

alginate-based formulations enriched with lemongrass essential oil showed antimicrobial activity against several fungi [6] and were able to preserve the firmness and colour of 'Rocha' pears [7]. Moreover, extracts of olive leaves loaded into alginate microbeads formed edible matrices with improved thermal stability, as detected by differential scanning calorimetry (DSC), and scanning electron microscopy (SEM) [8]. Also, mangoes packed into multilayer coatings composed of cinnamon essential oils and alginate, were able to preserve the firmness, edible content, and commercial value of the fruits, after 14 days, at 25 °C [9]; while edible coatings containing oregano essential oil improved the shelf life of tomatoes, by reducing endogenous microbial growth over 14 days, at room temperature [10].

In what concerns tissue engineering, calcium enriched alginate hydrogels were able to promote neovascularization and osteochondral repair [11]. In addition, Pereira et al. showed that *Aloe vera* enriched alginate films presented suitable properties for wound healing and drug delivery applications [1, 12]. In what respects bioremediation for instance, an alginate-coated magnetic nanocluster with high adsorption capacity and high selectivity for inorganic mercury has been developed as a nanosorbent tool for wastewater treatment [13]. Thus, given their versatility and sustainability, the use of alginate-based enriched biomaterials continues to grow and find new applications among several fields.

1.2 Pectin-Based Enriched Biofilms

Pectins are plant cell wall polysaccharides mainly composed by galacturonic acid [14]. Depending on composition, structure, and molecular weight, pectins can be classified in homogalacturonans, rhamnogalacturonans, xylogalacturonans, arabinogalactans and arabinans [14]. Pectins can undergo gelation and work as emulsifying, gelling, stabilizing and thickening agents, with several uses amongst various fields, given their low cost, non-toxicity, high stability, and biocompatibility [14–22].

Edible pectin films hold good mechanical and barrier properties to apply in the coating or packaging of fruits and vegetables [14, 15]. Pectin films enriched with blackcurrant pomace powder, for instance, were used to minimize food production losses and to increase the functional properties of food coatings and wrappers [16]. In addition, edible pectin coatings enriched with clove essential oil (CEO) were also able to act as seafood preservatives [17]. These CEO pectin-based films inhibited bacterial growth; improved the water holding capacity; prevented lipid oxidation and extended the shelf life and textural attributes of bream fillets, by at least 15 days, under refrigeration [17]. Moreover, pectin-based biomaterials also hold great potential for biomedical applications, given their natural prebiotic, dietetic, hypoglycemic, hypocholesterolemic, and anti-cancer effects [18–20]. In what regards bioremediation, pectin-based materials have been applied with success to pollutants removal due to their special ability to coordinate divalent cations [21, 22]. Commercial citrus pectin, sugar beet pulp pectin and sweet potato pectin have been able to remove heavy metals (Pb(II), Cu(II), Co(II) and Zn(II)) and organic dyes from aqueous solution [21, 22].

1.3 Cellulose-Based Enriched Biofilms

Cellulose is a natural polymer of D-glucose connected by $\beta(1 \rightarrow 4)$ glycosidic bonds [23, 24]. Cellulose is present in all plants to provide for structure and mechanical integrity. Cellulose fibers are highly hydrophilic, have low density, are nonabrasive and can be used as natural thickeners, binders, emulsifiers, suspending agents, surfactants, lubricants, stabilizers, and additives [23, 24]. Guava fruits coated with cellulose films, for instance, showed increased firmness, color maintenance, and reduced ripeness, for 13 days, at room temperature [25]. Additionally, cellulose-based films impregnated with esters of caffeic acid were able to show increased antioxidant activity and antimicrobial effects against various microorganisms [26]; while cellulose-based composites containing natural antimicrobials were able to inhibit the growth of food spoilage and pathogenic microorganisms such as Listeria monocytogenes, as well as to enhance the safety and quality of cold smoked salmon [27] and of individually packaged hot dogs [28]. Moreover, cellulose nanoparticles showed enhanced antioxidant activities of curcumin and ascorbyl dipalmitate when employed in packaging films [29]; and showed antimicrobial activity when applied to mature-green tomatoes (inhibition of Salmonella Montevideo and maintenance of color and firmness) stored at 20 °C, for 18 days [30]. Furthermore, cellulose-based-nanomaterials have also been used for tissue engineering, wound dressing, and 3D bioprinting [23, 24, 31], as well as for bioremediation and water treatment, by membrane filtration, catalytic degradation, absorption, adsorption, and/or disinfection [32-34]. Cellulose-based hydrogels formed by sonochemistry, for instance, use green-chemistry processes (with reduced reaction steps and chemical reagents), that also promote biocompatibility [35].

2 Conclusions

A sustainable and recycle-based society can be developed through the use of renewable and bio-based materials. Multifunctional alginate-based, pectin-based, and cellulosebased enriched structures can be applied to several industrial fields. These natural materials are key to promote environmental sustainability and continue to amaze by their properties and advantages, either presented separately or as conjugated biocomposites.

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