



Review

A review on synthesis, properties and applications of natural polymer based carrageenan blends and composites



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ABSTRACT

Carrageenan is a natural polysaccharide extracted from edible red seaweeds of Rhodophyceae class. It has been used as a viscosity increasing or gelling agent for prolonged and controlled drug release, food, pharmaceuticals and other industries. However, in spite of wide range of applications, carrageenan has some drawbacks and adverse effects on the biological systems, so its modifications with natural and synthetic polymers are carried out. This review article presents different sources and properties of carrageenans with special emphasis on natural polymer based carrageenan blends and composites and their applications in controlled drug delivery system, wound dressing and tissue engineering because of their biodegradability and biocompatibility, food industry as thickening/gelling materials, cosmeceuticals and making polyelectrolyte complexes.

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Abbreviations: AFM, atomic force microscopy; CA, contact angle; CLSM, confocal laser scanning microscopy; CMC, carboxy methyl cellulose; DLS, dynamic light scattering; DMA, dynamic mechanical analyses; DSC, differential scanning calorimetry; EIS, electrochemical impedance spectroscopy; EMC, equilibrium moisture content; FESEM, field-emission scanning electron microscopy; FTIR, fourier transform infrared; HPLC, high performance liquid chromatography; IEC, ionic exchange capacity; IR, infrared spectroscopy; LDV, laser doppler velocimetry; LSW, linear sweep voltammetry; MTT, methyl thiazolyl tetrazolium; NMR, nuclear magnetic resonance spectroscopy; SEM, scanning electron microscopy; TEM, transmission electron microscopy; TGA, Thermogravimetric analyses; TOF-SIMS, time of flight secondary ion mass spectrometry; WCA, Water contact angle; WPI, whey protein isolate; WRV, water retention values; WVP, water vapour permeability; XPS, X-ray photoelectron spectroscopy technique; XRD, X-ray diffraction.

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1. Introduction

Algae are photosynthetic organisms with a complex and controversial taxonomy. There are two main types of algae: (i) microalgae, the little size algae, which are found in benthic and littoral environments and throughout the sea waters as phytoplankton and (ii) macro algae (seaweeds), which occupy the littoral zone and are of large size [1,2]. Seaweed is a multicellular photosynthetic microorganism and potential biomass resource for the full or partial substitution and displacement of terrestrial biomass to generate valuable biochemical products and biofuels [3]. Algal biomass is an excellent candidate for renewable sources due to specific characteristics such as higher growth rate, high photosynthetic productivity, extraordinary potential for carbon dioxide fixation [4], high amount of carbohydrates and lower amount of lignin [5,6].

Seaweed algae have large amount of cell wall polysaccharides, storage polysaccharides and mycopolysaccharides [7]. Algal polysaccharides are gaining interest due to their sustainability and availability in large amount and their unusual chemical composition which is not present in any other organisms [8]. Algal polysaccharides are a wide class of biopolymers that are derived from green seaweeds such as ulvans, brown seaweeds such as alginates and red seaweeds such as carrageenans. Carrageenan and alginates due to their specific properties i.e. (thickening & gelling agents) and their biological activities (antiviral, immune-inflammatory and anticoagulant) are mainly used in pharmaceuticals, cosmetics and food industry [9].

Carrageenan (CG) is the general name for a group of high molecular weight sulphated polysaccharides obtained by extraction of red seaweeds formed by alternate units of D-galactose and 3, 6-anhydro-galactose (3, 6-AG) joined by α -1, 3 and β -1, 4-glycosidic linkage [10–12]. CG is a sulfated polygalactan with 15–40% ester-sulfate content, which makes it an anionic polysaccharide. CG is used as a gelling, stabilizing and thickening agent and also as fat substitutes in food industry, especially in milk products [13]. They are also used in different non-food items, for example, cosmetics [14], pharmaceutical [15], textile formulations and printings [16].

1.1. Sources

Carrageenan is extracted from red seaweed of the Rhodophyceae family [17–24], commonly from genera such as *Eucheuma*, *Solieria*, *Cripus*, *Agardhiella*, *Chondrus*, *Hypnea*, *Sarcocystis*, *Iridaea*, *Gigartina*, *Gigartina* and *Agardhiella* [25–39]. *Eucheuma* and *Kappaphycus* seaweeds are most commonly cultivated seaweed across Malaysia and Southeast Asia [40].

1.2. Structure and physicochemical properties of carrageenans

Carrageenans are linear sulfated polysaccharides composed of repeating disaccharide units with alternating 3-linked β -D-galactopyranose and 4-linked α -galactopyranose or 3, 6-anhydro- α -galactopyranose (Fig. 1) [41,42].

1.2.1. Types of carrageenan

There are three main types of carrageenan, which vary in their degree of sulfation [14,15,24,43]. (i) **Kappa (κ)-carrageenan** is composed of alternating 3-linked β -D-galactose 4-sulfate and 4-linked 6-anhydro- α -galactopyranose having one negative charge per disaccharide repeating unit (Fig. 1a) [41,44]. (ii) **Iota (ι)-carrageenan** has two sulfate groups per disaccharide repeating unit (Fig. 1b) [41,44]. Iota and kappa carrageenan show similar properties such as, κ - and ι -carrageenan in aqueous solution undergoes a thermo reversible conformational arrangement at higher temperatures while at lower temperatures they undergo combination and network formation through sulphate groups and the 3,6-anhydro-D-galactopyransyl ring [45–47]. ι and κ -carrageenans also exhibit gelling properties in the presence of cations which is influenced by the concentration and the nature of cations (i.e. K^+ and Ca^{2+} , respectively) present in the solution as well as by the biopolymer concentration [45,47]. Moreover, several types of salts have different effects on the phase transitions and gelling of ι - and κ -carrageenan gels. It has been reported that stronger κ -carrageenan gels were formed in the presence of KCl as compared to the other salts i.e. NaCl, $MgCl_2$, LiCl, $SrCl_2$ and $CaCl_2$, [48]. Meanwhile, the gel-sol transition temperatures of κ -carrageenan were strongly related to KCl, NaCl, and $CaCl_2$ contents [49]. For ι -carrageenan storage modulus is increased rapidly with the divalent salt concentration and slowly with the monovalent salt concentration. In contrast, for κ -carrageenan storage modulus is increased more rapidly in the presence of KCl than that with copper or calcium. However, for high salt concentrations storage modulus became independent of the concentration and type of cations in the κ -carrageenan solution [50]. (iii) **Lambda (λ)-carrageenan** has three sulfate groups per disaccharide unit but they do not exhibit any 3, 6-anhydride bridge contrary to κ and ι -carrageenan (Fig. 1c) [41,44]. λ -carrageenan is unable to form gel and has a random coil conformation at all temperatures [45]. A recent research showed that gelation in λ -carrageenan certainly was possible with trivalent ions. This novel finding has potential to increase λ -carrageenan present uses other than a viscosity increasing agent [51].

The main differences which effect the properties of different carrageenans are the number and position of ester sulfate groups and the content of 3,6-anhydro-galactose (3,6-AG). Higher levels

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