Renewable and Sustainable Energy Reviews 77 (2017) 353–362

Contents lists available at ScienceDirect

Renewable and Sustainable Energy Reviews

journal homepage: www.elsevier.com/locate/rser

Seaweed based sustainable films and composites for food and pharmaceutical applications: A review

H.P.S. Abdul Khalil[a,*], Chaturbhuj K. Saurabh[a], Y.Y. Tye[a], T.K. Lai[a], A.M. Easa[a], E. Rosamah[b], M.R.N. Fazita[a], M.I. Syakir[a], A.S. Adnan[c], H.M. Fizree[a], N.A.S. Aprilia[d], Aparajita Banerjee[e]

[a] School of Industrial Technology, Universiti Sains Malaysia, 11800 Penang, Malaysia
[b] Faculty of Forestry, Mulawarman University, Samarinda, East Kalimantan, Indonesia
[c] School of Medical Sciences, Universiti Sains Malaysia, 11800 Penang, Malaysia
[d] Department of Chemical Engineering, Syiah Kuala University, Banda Aceh, Indonesia
[e] Department of Biotechnology & Microbiology, Tilak College of Science & Commerce, University of Mumbai, 400032 Mumbai, India

ARTICLE INFO

Keywords:
Seaweed
Alginate
Carrageenan
Agar
Nanomaterials
Essential oils

ABSTRACT

Various studies have been focused on seaweeds derived polysaccharides based composites because of its renewability and sustainability for food packaging and pharmaceutical applications including tissue engineering, drug delivery, and wound dressing. Alginate, carrageenan, and agar are widely used for this purpose due to their biocompatibility, availability, gelling capacity, and encapsulation efficiency. Essential oils (like oregano, clove, lemongrass, etc.) as antimicrobial and antioxidant agent, biopolymer (like starch, cellulose, chitosan, etc.), and nanoparticles (organically modified and unmodified inorganic nanoclays, nano-cellulose, carbon nanotubes) as reinforcing material are frequently used for the fabrication of seaweed based materials. Composites have an edge over pure polymer based material in terms of mechanical and barrier properties, controlled release of drugs, and adsorption efficiency. This review comprehensively addresses different types of additives and their impact on various functional properties of seaweed based composites, their methods of incorporation, and applications with special emphasis on food and pharmaceutical usage.

1. Introduction

Packaging dominates the waste generated from plastics. Major hurdles against increasing use of plastics are non-biodegradability and derivability from non-renewable natural resources. This has put tremendous pressure on the environment due to the accumulation of plastic products in natural surroundings which adversely affect wildlife, wildlife habitat, and humans. Thus there is need to derive packaging for novel polymers to address the shortcomings of conventional plastics. Biopolymers such as starch, gluten, and guar gum are suitable alternatives to fabricate packaging material due to their nontoxicity, biodegradability, and derivability from renewable natural resources. Seaweed based polysaccharides are an interesting example of biopolymer and films derived from such source have good oxygen vapor barrier properties and are impervious to fats and oils. However, the major limitations in the use of biopolymers as packaging materials are their relatively poor mechanical and barrier properties as compared to their nonbiodegradable counterparts [1]. One of the most frequently used methods to overcome this drawback is to fabricate composite films by mixing of one polymer with another polymer and/or hydrophobic component and/or nanoparticles. This approach enables one to utilize the distinct functional characteristics of every component of the composite film. Thus such hybrid films have improved mechanical and barrier properties over the pure polymeric film [2,3].

For the development of food packaging when seaweed based polysaccharides films reinforced with organically modified or unmodified nanoclay an improvement in mechanical strength of the film was observed [4,5]. Moreover, strong inhibitory activity against foodborne pathogens in seaweed based food packaging is developed by incorporating natural or synthetic antimicrobial agents like grapefruit seed extracts, silver nanoparticles etc. [6,7]. Through the vast literature survey, it can be concluded that additives like nonmaterial and antimicrobial component effectively improves various properties of composite films. Besides food packaging, seaweed based composites are also studied for pharmaceutical applications owing to its excellent properties.

* Corresponding author.
E-mail addresses: akhalilhps@gmail.com (H.P.S. Abdul Khalil), chaturbhuj_biotech@yahoo.co.in (C.K. Saurabh), tyying87@yahoo.com (Y.Y. Tye), tzekiat25@gmail.com (T.K. Lai), azhar@usm.my (A.M. Easa), enihros@yahoo.com (E. Rosamah), fazita@usm.my (M.R.N. Fazita), misyakir@usm.my (M.I. Syakir), drazreen@usm.my (A.S. Adnan), eg_fzzzry@yahoo.com (H.M. Fizree), sriaprilia@yahoo.com (N.A.S. Aprilia), banerjeeaparajita01@gmail.com (A. Banerjee).

http://dx.doi.org/10.1016/j.rser.2017.04.025
Received 7 September 2016; Received in revised form 24 January 2017; Accepted 17 April 2017
1364-0321/

In recent years pharmaceutical firms have started looking towards marine organisms including seaweeds for novel drugs delivery systems from natural products. Due to excellent hydrogel-forming capability under relative pH [8], biocompatibility, and hypoallergenic nature seaweed offer a broader platform for medicinal purpose especially in drug delivery and tissue engineering. Seaweed polysaccharides have hydrophilic groups like carboxyl, sulfate, and hydroxyl groups on the surface which can easily interact with biological tissues. Sulfated polysaccharides have anionic sulfate groups which are not present in polysaccharides of terrestrial and animal origin. Such polysaccharides avoid aggregation during blood circulation by reduced interaction with serum proteins. Owing to these properties the usage of seaweed polysaccharides in biomedical applications are increasing. However, due to poor mechanical strength, there is limited use of pure seaweed derived polysaccharides in pharmacy including bone tissue engineering. Poor functional characteristics of seaweed polymers also lead to rapid release of drugs and loss of bioactive compounds under formulating environments such as heat, sonication, or exposure to organic solvents. Various studies have shown that composite materials can effectively address such issues, for example, nanoclay or biopolymer addition in seaweed matrix resulted in a sustained release of drugs [9,10]. Herbal or biopolymer incorporated seaweed scaffolds had improved cell adhesion and proliferation as compared to scaffolds without herbal extract or biopolymer [11,12]. Thus we can conclude that seaweed based composites have many added advantage over pure biopolymer-based material in various pharmaceutical applications.

Although both food and pharma sector have parallels between their overall production practices, they also have their own discrete challenges. Extensive reviews are available on food and pharma applications of biopolymers like starch, cellulose, polylactic acid (PLA), polycaprolactone (PCL), etc. However, comprehensive review on seaweed for food packaging and pharmaceutical applications is lacking. Thus there is a need to provide an insight on this with current trends and future prospect. The objective of this review is to discuss comprehensive scenario about the types and properties of seaweed based composites along with their preparation procedure and applications with special emphasis on pharmaceuticals and food packaging.

1.1. Chronological event in the field of seaweed based composites

Humans use seaweeds since the time immemorial and date back to some of the earliest records in human history (Table 1). Agar was discovered around 1658 in Japan and its first chemical analysis was done in 1859. Carrageenan was used as food additives since the 15th century. However, as an industrial crop, seaweed cultivation is still a recent development following its rapid increase in production and technological developments during the past half century. Seaweed as a crop has established itself as one of the most transitional industries with rapid development potential that can address the long-term issue of environmental sustainability.

1.2. Seaweed properties and extraction for food packaging and pharmaceuticals

Seaweed refers to several species of macroscopic, multicellular, marine algae that live near the seabed. The term includes some members of the red, brown, and green algae. They are the most abundant source of polysaccharides including alginate, agar, fucoidan, agarose, carrageenan, and ulvan. Agar is derived from a polysaccharide called agarose, which forms the supporting structure in the cell walls of red algae of Rhodophyceae class [23]. Agarose is accountable for the gelling capacity of agar which makes it very useful in skin care, herbal medicines, and has excellent film properties. Due to this agar and agar-based composites are widely used in food and pharmaceutical applications [5]. Most of the large brown seaweeds are potential sources of alginate. Alginic acid is a linear copolymer with homopolymeric blocks of (1-4)-linked β-D-mannuronate and its C-5 epimer α-L-guluronate residues, respectively, covalently linked together in different sequences. It has been shown that the physical properties of alginates depend on the sequencing of its monomers [24]. The ability of alginates to react with divalent and trivalent cations is widely being utilized in alginate film formation. Alginic acid is insoluble in water but swells when placed in water. This property makes it a useful disintegrating agent in tablets. Carrageenans are linear sulfated polysaccharides that are extracted from red edible seaweeds. It has three main commercial classes: Kappa (κ) forms strong, rigid gels in the presence of potassium ions and due to such properties it has been used for the formation of cohesive and transparent films [25]. Iota (ι) forms soft gels in the presence of calcium ions. Lambda (λ) does not form the gel and is used to thicken dairy products. Carrageenans are widely exploited due to its hydrophilic and anionic properties. It has been extensively used in food packaging and pharmaceutical industries as gelling, emulsifying, stabilizing agents, and base material for packaging films. According to FAO's report on Global Aquaculture Production published in 2013, the worldwide production of seaweed was around 27 million tons (Table 2).

Seaweeds are mainly harvested for human consumption. Besides that, they are also being cultivated for the extraction of gelatinous substances collectively known as hydrocolloids: alginate, agar, and carrageenan. Alkali is used for the extraction of carrageenans because it induces chemical modification that leads to increased gel strength in the final product. There are two methods for recovering carrageenans

Table 1
Chronological event of seaweed usage and recent technological development.

Period	Stage of development in applied seaweed research and industry
≥35,000 BCE	Water carriers were made using hygroscopic properties of kelp by Tasmanian aboriginal (Dillehay et al. [13]).
13,000 BCE	Ancient civilizations in Chile used seaweed for nutrition and health (Dillehay et al. [13]).
0–300 CE	In China it was used for iodine supplement (Dillehay et al. [13]).
1716	Alginate was reported to be used in wound dressings (Martin [14]).
1940s	Industrial development of hydrocolloids from seaweeds; research on seaweed as food (Craigie [15]).
1948	Incorporation of antioxidants to the carrageenan based coatings for improved quality and microbiological stability of muscle foods (Stoloff et al. [16]).
1950s	First international seaweed symposium on seaweed held at Scotland (Dillehay et al. [13]).
1961	Calcium alginate coatings reduced dehydration of cut-up poultry (Mountney and Winter [17]).
2000	Inhibition of *Salmonella* on broiler skin using agar based films containing nisin (Natrajan and Sheldon [18]).
2010	Development of chitosan/carrageenan composite nanoparticles for drug delivery applications (Grenha et al. [19]).
2012	Alginate based nanocomposite film reinforced with nanocrystalline cellulose was prepared (Huq et al. [20]).
2013	Agar film incorporated with silver nanoparticles exhibited improved water vapor, gas barrier and mechanical properties (Rhim et al. [7]).
2014	Carrageenan based composite films incorporated with grape seed extract showed strong inhibitory activity against food borne pathogens (Kanmani and Rhim [6]).
2015	Alginate/pullulan based composite films were developed (Xiao et al. [21]).
2016	Nanocomposites were prepared using cellulose nanofibrils and alginate biopolymer (Deepa et al. [22]).