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REVIEW PAPER

Seaweed biorefinery

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Abstract Seaweed offers a wide range of interesting bioactives. With over 10,000 species globally, it is of great interest to be able to extract these compounds. Hence, seaweed fractionation into a wide spectrum of valuable products using multistage cascade processes offers a sustainable approach of exploitation of this resource to produce bioactive ingredients, chemicals and biofuels. This biorefinery processing approach should be adapted to local conditions to maximize the biomass utilization and to lower the waste fractions or preventing any waste materials re-enforcing the circular economy. This review presents an overview of the potential uses of waste generated after seaweed processing for food and other uses, as well as the utilization of invasive species biomass and other invasive species.

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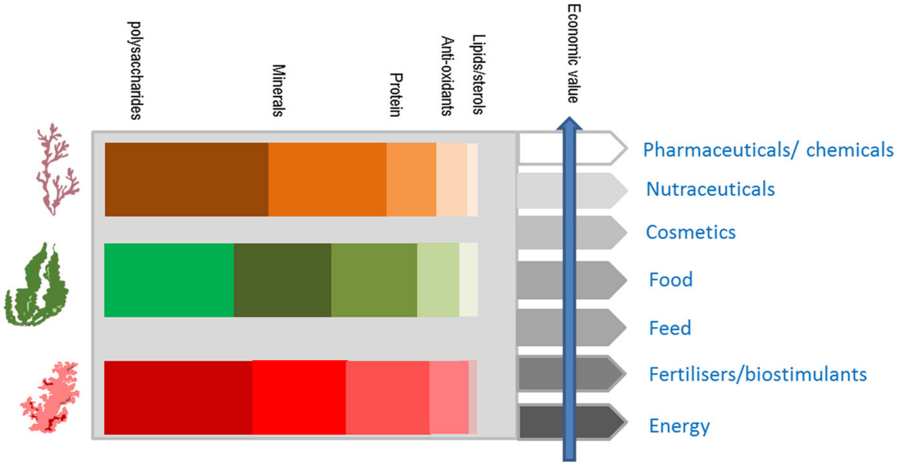
Graphical abstract



Keywords Seaweed · Biorefinery · Food · Feed · Cosmetics · Pharmaceuticals · Fertilizer · Energy

Abbreviations

ABTS	2,2′-Azino-bis(3-ethylbenzothiazoline-6-sulphonic acid)
ACE	Angiotensin-converting-enzyme
ALT	Alanine aminotransferase
AST	Aspartate aminotransferase
BWG	Body weight gain
CF	Crude fiber
CP	Crude protein
d.w.	Dry weight
DCP	Digestible crude protein
DE	Digestible extract
DM	Dry matter
DNA	Deoxyribonucleic acid
DPPH	2,2-Diphenyl-1-picrylhydrazyl
EE	Ether extract
FCM	Fat corrected milk
FI	Feed intake
HaCaT cells	Spontaneously immortalized human keratinocyte line
MAAs	Microsporyne amino acids
MMP-1	Fibroblast collagenase
NFE	Nitrogen free extract
NSP	Non-starch polysaccharides
OM	Organic matter
SHF	Separate enzyme hydrolysis and fermentation
SNF	Solids non fat
SSF	Simultaneous saccharification and fermentation
SWOT	Strengths, weaknesses opportunities, threats
TDN	Total digestible nutrient
TS	Total soluble
UV	Ultraviolet
UVB	Ultraviolet B radiation
V	Volume
VS	Volatile solids
wt	Weight

1 Introduction

Seaweeds have traditionally been used for food applications, as whole sea vegetable or for the extraction of phycocolloids in the food processing industry (agars, carrageenan and alginates). Furthermore, they have been used for many centuries as animal feed ingredient and fertilizers in agriculture and horticulture (Kraan and Guiry 2006). Seaweeds are a source of biologically active phytochemicals including pigments, fatty acids, polysaccharides, vitamins, minerals, sterols, terpenoids etc. (Dominguez 2013), showing biological properties with potential benefits in a range of diseases such as, control of hyperlipidemia, thrombosis, tumor, and obesity (Kim 2011). Seaweeds can also be used primarily as substantial feedstock for biomass, biofuel production, pharmaceutical and cosmetic sectors (Kraan 2013; Mahadevan 2015; Tiwari and Troy 2015; Barbot et al. 2016). To make use of all seaweed components, with their functionalities and bioactivities intact requires a rational processing of the whole material, and the algal processing by-products (Samarakoon et al. 2014).

The biorefinery concept has been proposed in order to utilize all valuable seaweed constituents in an economically feasible cascading process adopting a zero waste approach while reducing impact on climate change (Kumar and Sahoo 2017; Schiener et al. 2016a, b; Kostas et al. 2017). Biorefinery has also been described as a biomass deconstruction process allowing to get the full potential of biomass (Baghel et al. 2016) and is assuming significant importance in the context of a marine sustainable bio-economy (Gajaria et al. 2017). The necessity of sustainable utilization of seaweeds and effective management of these resources by coastal communities has been highlighted (Mac Monagail et al. 2017).

Over the last decade, the increasing worldwide demand of seaweed has stimulated aquaculture activities outside of the traditional seaweed farming countries such as Korea, Indonesia, China and Japan, resulting in an increased seaweed waste stream from industrial processes, either as rejected material before processing when quality standards are not meet or as left over from the processing industry (Quitain 2013). Non-food uses can rely on the utilization of residual and waste biomass avoiding competition with seaweed demand from the food industry (Hughes et al. 2012; Schiener et al. 2017). Furthermore, in order to use algal biomass effectively and decrease cost of the algal processes, research on utilization of algal wastes for different purposes are examined (Özçimen et al. 2015). The disposal and reutilization of seaweed waste is essential for the preservation of the marine environment and recycling of organic substances (Kim et al. 2013a, b) contributing to the circular economy concept instead of the linear model of make, use and waste. Traditionally waste materials have been collected and often used as feed, indicating they are under-utilized (Ferrero et al. 2015). Another application for algal waste is the use in in agricultural and composting, but low demand has little added value to the collected seaweed wastes (Hardouin et al. 2014a, b). Seaweed sources are not limited to cultivated seaweeds. Seaweed processing waste possesses several biological activities of interest for various pharmaceutical and nutraceutical applications. Integrated biorefinery solutions have sufficient scale to enable the economic production of fuel from underutilized seaweeds or from waste fractions (Tiwari and Troy 2015).

During the last 10 years, there is a marked increase of proliferations of green, brown and red seaweeds, caused most probably by eutrophication in combination with climate change. Green tides, red tides and brown tides are nowadays a common phenomenon and appear on beaches to represent a nuisance for residents and tourist activities. Collecting of this nuisance seaweed biomass will take nutrients like phosphate and nitrates out of the ecosystem and hence may prevent further eutrophication processes and damages to marine ecosystems with a positive impact on local tourism. The collected biomass could serve e.g., as biogas feedstock material. Currently seaweed green or brown tides represents an underutilized biomass feedstock. Recurrent blooming events can cause serious environmental and economic problems (Wang and Hu 2017). Research projects have been initiated to predict these bloom events. Removal in hypertrophication-affected areas would greatly contribute to the improvement of environmental conditions for beaches and marine areas, improving management for local tourism (Barbot et al. 2016).

As a raw material, cultivated seaweeds offer important advantages in terms of sustainability, as: (1) cultivation in the sea does not require arable land and freshwater inputs and requires minimal human intervention, (2) fertilizers are not needed since nutrients are absorbed from the surrounding water, (3) seaweed present a rapid reproduction rate and offer high biomass yield, (4) the fast growth rate helps to reduce atmospheric greenhouse carbon through photosynthesis and supply oxygen to the sea acting as a short term carbon sink and help to alleviate ocean acidification (Kraan 2013). Invasive species and biomass from seaweeds bloom represent an economic constraint for the affected communities, hence their valorization is an opportunity for producing new compounds of interest (Goh and Lee 2010; Hardouin et al. 2014a, b; Mahadevan

2015; Kositkanawuth et al. 2017; Balina et al. 2013). Offshore seaweed production for nutrient circular management is an instrument to achieve better water quality (Seghetta et al. 2016a, b, c). However, there are significant challenges associated with the development of seaweed-based ingredients due to varying levels of bioactive compounds in the species and seasonal factors. (Hafting et al. 2015; Balboa et al. 2016; Tedesco and Daniels 2018). In contrast, industrial residues are generated with continuous amounts and similar biomass composition, and easily linked to the production process. Seaweed biomass from eutrophication shows a much higher variation in composition and a high fluctuation in availability, requiring biomass storage to assure a steady supply of biomass.

There are comprehensive reviews on seaweed biorefinery process and compounds and bioactives of interest for seaweeds from European temperate Atlantic waters (Holdt and Kraan 2011). Cultivation, production, chemical composition and their potential added-value products and applications (Jung et al. 2013; Lorbeer et al. 2013; Winberg et al. 2014), the production of renewable chemicals, fuels and materials employing green chemical technologies (Kerton et al. 2013), the conversion potential of algal industrial wastes (Özçimen et al. 2015) and the use of seaweeds from industrial residues or eutrophication as an alternative feedstock for biofuels (Fernand et al. 2017) and biogas production (Barbot et al. 2016). Moreover, Balina et al. (2017) describe the biorefinery concept and its implementation in bioeconomy through a SWOT analysis to reach the set targets of low-carbon economy, higher sustainability and optimal bioresource use efficiency. Nevertheless, most literature on the biorefinery approach is applied to microalgae. Some of the extraction and purification methods for compounds from microalgae are of interest for seaweed biomass. However, the composition and nature of seaweeds and the industrial existing processes for the extraction of other components (i.e. hydrocolloids) point to different extraction scenarios. This review provides an overview of the potential applications of seaweed bioactives from underutilized or invasive seaweed and seaweed blooms and the food processing waste. It summarizes the existing methods with conventional and novel technologies to extract and purify such metabolites in order to develop feasible and sustainable industries and illustrates the cascade valorization of these biorenewable sources.

2 Seaweed composition and properties

Seaweeds have a unique chemical composition mostly known for their polysaccharides or hydrocolloids such as agars, carrageenan and alginates, which are widely used in the food and many other industries (Bixler and Porse 2011). Coupled to their fast growth rate and biomass production seaweeds offer an interesting source for biorefining. Seaweeds contain nutritional elements such as proteins, lipids, carbohydrates, vitamins and minerals, the content of these elements varies depending on the season and the area of production (Connan et al. 2004; Khan et al. 2007; Marinho et al. 2016; Murata and Nakazoe 2001; Zubia et al. 2008). Over 50 different minerals and trace elements are present in seaweeds, which are required for the body's physiological functions (Kraan 2013). Seaweeds in general have high levels of macro elements such as, Potassium, Calcium, Iron, and Iodine. Furthermore, most vitamins are present except D vitamins and they contain high levels of various antioxidants (Holdt and Kraan 2011).

2.1 Minerals

It is a well-known fact that seaweeds are particularly rich in minerals and trace elements. Minerals such as Potassium, Calcium, Iron and Magnesium are present in seaweeds at much higher levels than many if not most land vegetables. Besides macro elements seaweeds are also a good source of the rarer trace elements such as Manganese or Cobalt (Kraan 2013).

The ash content in seaweed is high when compared to vegetables (Murata and Nakazoe 2001). This high level of ash, basically minerals and trace elements are attributed to their capacity to retain inorganic marine substances due to the characteristics of their cell surface polysaccharides (Mabeau and Fleurence 1993). All of the essential minerals and trace elements needed for human nutrition are present in seaweeds (Rupérez 2002) and indicate that seaweeds could be applied as functional food.

2.2 Carbohydrates

The structural polysaccharides (agar, carrageenan and alginate) in seaweeds can range up to 40–50% of the dry weight. Seaweed also contain various storage polysaccharides such as laminarin, fucoidan, porphyrans, floridian starch and ulvans (Holdt and Kraan 2011). Several of the sulphated polysaccharides have implication for human health and will be discussed elsewhere. These polysaccharides are generally not digested by humans and are regarded as dietary fibers (Mabeau and Fleurence 1993). MacArtain et al. (2007) showed that seaweeds are a good source of dietary fiber compared to other terrestrial foodstuffs. Consuming seaweeds can especially help with gut health and associated issues for health and well-being. These polysaccharides providing soothing action, adding bulk to the digestive tract and lowering cholesterol uptake (Lahaye 1991; Mendis and Kim 2011).

2.3 Proteins, peptides and amino acids

Protein and the amino acid composition of seaweeds vary among different algal groups and it is found to be highest among red seaweeds, followed by green and brown seaweeds. Generally, the brown seaweeds contain protein as low as 3–15% of dry weight compared to green or red seaweeds having 25 up to $>$ 40% protein of dry weight. The protein content of *Porphyra* and *Palmaria* can be compared with high protein soybean (Fleurence et al. 1999). Algal amino acid composition has been frequently studied and the dominant forms are aspartic and glutamic acids, which is of interest as an umami factor (Hotchkiss 2012; Qi 2017). Other amino acids, such as lysine, leucine, etc., are also present in substantial concentrations. The high percentage of protein and amino acids in seaweeds makes them an interesting potential source for biorefinery processes (Murata and Nakazoe 2001). Habitat and especially seasonal variation, have an effect on protein, peptides and amino acids in seaweed (Arasaki and Arasaki 1983; Haug and Jensen 1983) and indicates the importance of time of harvest and geographical location.

2.4 Lipids and fatty acids

Lipids and fatty acids are present in low amounts, generally 2–4% of the dry weight in seaweeds (Khotimchenko 2003; Holdt and Kraan 2011) although they contain relatively higher levels or polyunsaturated fatty acids (such as omega 3 and omega 6 acids) than vegetables (Mendis and Kim 2011). Polyunsaturated fatty acids are known to have various roles in the prevention of cardiovascular diseases and provide other health benefits and will be discussed elsewhere. Seaweeds also contain various other lipids and lipid like compounds such as sterols (Sánchez-Machado et al. 2004; Whittaker et al. 2000), terpenoids and tocopherols (Haugan and Liaaen-Jensen 1989); phospholipids in red seaweeds (Dembitsky and Rozentsvet 1990) and in brown seaweeds (Jones and Harwood 1992) and glycolipids (Dembitsky et al. 1990, 1991).

2.5 Vitamins

Although not a rich source, seaweeds do contain both water-and fat-soluble vitamins such as A, most B vitamins and particularly B12, C, and E. Some species also carry Vitamin H and K. The brown seaweeds Ascophyllum and Fucus contain higher levels of vitamin E than green and red seaweeds. Brown seaweeds contain α-, β-, and γ tocopherol while green and red seaweeds contain only α-tocopherol. Vitamin B12, which is generally present in animal products and is particularly recommended to mitigate the effects of aging and anemia, is also present in many seaweeds including *Porphyria*, Ulva, Ascophyllum, Laminaria and Palmaria. The source of the vitamin B12 in seaweeds is proposed to be bacteria living on the surface or in the adjacent waters.

2.6 Pigments

Three basic classes of pigments are found in seaweeds, i.e. chlorophylls, carotenoids and phycobilliproteins. Under these categories many pigments have been described in seaweeds all with their own specific bioactivities and health effects (Holdt and Kraan 2011). These pigments can act as strong anti-oxidant. By helping to protect tissues against oxidative stress, pigments will have a positive effect for problems such as cardiovascular diseases, cancers, arthritis, and autoimmune disorders. Some have also exhibited anti-inflammatory and hepatoprotective effects (Maliakal et al. 2001; Garbisa et al. 2001). Carotenoids is the largest group of these pigments. Several studies have shown the correlation between a diet rich in carotenoids and a diminishing risk of cardio-vascular disease, cancers (β-carotene, lycopene), as well as ophthalmological diseases (lutein, zeaxanthin). Brown seaweeds are particularly rich in carotenoids especially in fucoxanthin, β-carotene, violaxanthin. It has been demonstrated that fucoxanthin inhibits the proliferation of HL-60 cells (human leukemia cell line) and induces their apoptosis (Hosokawa et al. 1999). The main carotenoids in the red seaweeds are the β-carotene and α-carotene and their dihydroxylated derivatives: zeaxanthin and lutein. The carotenoid composition of the green seaweeds is similar to that of higher plants: the main carotenoids present are the B-carotene, lutein, violaxanthin, antheraxanthin, zeaxanthin and neoxanthin. Several studies demonstrated the antioxidant properties of the algal carotenoids and the role they play in preventing many pathologies linked to oxidative stress (Okuzumi et al. 1993; Yan et al. 1999).

3 Uses

Traditionally the dominant markets for seaweeds are the food (whole seaweed and phycocolloids) and feed industry, but algal biomass are a renewable source for various products useful for agricultural, bioplastics, dyes, cosmetic, pharmaceutical, energy and bioremediation applications (Holdt and Kraan 2011; Cardozo et al. 2007).

A survey on the uses of waste generated after utilization of seaweeds for food uses is presented here, including the low quality fractions removed before direct consumption of the seaweeds, the solid residues discarded after the extraction of phycocolloids, as well as the utilization of invasive species biomass and the biomass from algal blooms and offshore arrivals.

Seaweed processing consist of harvesting the seaweed biomass, either manual or by mechanical means and further stabilizing the biomass through drying of the seaweeds using traditionally direct sunlight and wind drying or drum or convection dryers using hot air. Low-cost methods of preservation like solar drying or ensiling can address the discontinuity of biomass problems (Milledge and Harvey 2016a, b). Other methods of drying such as freeze drying are not economically feasible for large scale biomass. Drying techniques have different effects on the nutritional, functional, and biological properties of seaweed. To reduce drying costs and to improve drying efficiency novel technologies may be employed to reduce the energy consumption without compromising the quality and nutritional aspects of seaweed, including ultrasonication, microwaves, and osmosis (Adam et al. 2015).

3.1 Food

Many seaweeds are edible and contain important nutrients such as proteins and amino acids, essential fatty acids, such as omega-3 fatty acids, vitamins, and minerals necessary for human growth, development and prevention of diseases (Gómez-Ordóñez et al. 2012). The popularity of seaweed-based foods also in countries without traditional consumption is on the increase. Seaweed is seen and recognized by the market as a superfood. Moreover, the attraction and interests for exotic and different cuisines and to the demand for healthy foods, either as a whole food (Mahadevan 2015; Bocanegra et al. 2009) or fermented products (Uchida and Miyoshi 2013), as novel supplemented products with functional properties (López-López et al. 2009; D'Arrigo et al. 2011; Schultz Moreira et al. 2013; Charoensiddhi et al. 2017; Admassu et al. 2018) or as replacers of other non-desirable ingredients, such as preservatives and colorants (Sellimi et al. 2017) is driving the seaweed or sea vegetable market. Fortification with seaweeds of cereal based extruded products has been proposed, i.e. an extruded product fortified with Sargassum tenerrimum showed enhanced nutritional and functional qualities of traditional extruded snacks (Singh et al. 2018).

This past year has seen market growth in United States and Europe of close to 30% in seaweed based snacks and is continue to do so as the snack industry adopts this ingredient. Coupled to large improvements in processing technology of seaweed form harvesting to drying and applications in different products it means that there are big- and so-far largely underdeveloped opportunities for new seaweed food product development. In the last 2 years a total of 44 new seaweed snack products were launched in the United States and 97 in Europe. Products range from seaweed cracker like snacks to seaweed salads, wraps, soups and burgers and even drinks such as soft drinks or craft beer (Sellimi et al. 2017; Singh et al. 2018). Seaweed's natural healthy stature will drive markets as people want to snack but snacking without guilt. Seaweeds can assist in this role as seaweed offers a low fat, protein and vitamin packed snack that has no bad associations. This will mean that the cultivation and harvesting of seaweeds will increase and offers increasing opportunities for seaweed biorefinery processes by using waste biomass from those processing industries.

3.2 Agriculture

Seaweed has traditionally been used as fertilizer for plants, due to their richness in minerals and trace elements. The high fiber content (polysaccharides) aid in moisture retention, making them an excellent soil conditioner. The seaweed extracts in liquid form are frequently used as plant fertilizers and growth stimulants, due to their content in auxins, cytokinins, polyamines, gibberellins, abscisic acid and ethylene. Moreover, phloroglucinol derivatives also could have an influence in plant growth (Mahadevan 2015; Nabti et al. 2017; Rengasamy et al. 2016a, b; Sasikala et al. 2016). Seaweed can be applied in granular and powder forms (Kumari et al. 2013) and in a novel micronutrient applications based on the seaweed biomass available (Tuhy et al. 2015).

Several application for extracts have been proposed from soaking the seeds (Sasikala et al. 2016; Renuka et al. 2013; Sujatha et al. 2013; Layek et al. 2016; Sarkar et al. 2018), as a foliar spray (Sasikala et al. 2016; Sabir 2015; Faissal et al. 2013; Stamatiadis et al. 2014; Babilie et al. 2015; Singh et al. 2015; Mahajan et al. 2016; Rengasamy et al. 2016a, b; Guerreiro et al. 2017; Layek et al. 2018), or by watering direct to the soil/compost (Tawfeeq et al. 2016). Some examples and benefits are in Table 1.

A frequently used liquid fertilizer is “sap”, the liquid phase obtained from mechanical pressing of the milled fresh seaweed after washing with fresh water. *Kappaphycus alvarezii* fresh biomass is widely used for this purpose, and once crushed yielded aprox. 0.7 m^3 t^{-1} fresh seaweed of a KCl rich juice rich and a granular biomass (0.04 t wt fresh seaweed (Mondal et al. 2013). The liquid phase can be clarified by centrifugation and supplemented with preservatives (Singh et al. 2016). Seaweed sap contains macro and micro nutrients, amino acids, vitamins and growth-promoting substances (Prasad et al. 2010). *Kappaphycus* sap (2.5–7.5%) sprayed on the foliage of the potato crop in combination with 50–100% of the recommended dose of fertilizers had a significant impact on the growth, yield and quality improvement of the crop, and at 7.5% can be used to substitute 25% of the recommended dose of fertilizer (Pramanick et al. 2017). Favorable effect of *K. alvarezii* sap applied as a foliar spray (5%), increased the yield of tomato fruit (61%), by increasing the number and size of fruit, their quality and macro and microelements content, probably due to an improved nutrient uptake by fruit and shoot. Furthermore, plants showed resistance to leaf curl, bacterial wilt and fruit borer (Zodape et al. 2011).

Applying these fertilizers will lower the carbon and phosphate footprint of mineral fertilizers. The combination of 5–7.5% sap applied with 100% recommended rate of fertilizers enhanced the grain productivity of maize and increased the P and K content (Singh et al. 2016). *Kappaphycus* and *Gracilaria* sap used at 5% improved the germination and seedling vigor and three foliar spray at 5 and 10% produced higher plant height, dry-matter accumulation, chlorophyll content and lower leaf temperature as compared to the control water spray (Layek et al. 2016). *Kappaphycus* and *Gracilaria* saps used at 10% on *Vigna radiata* showed beneficial effects on growth, yield, quality of grains in terms of protein, phosphorous and potassium content and also influenced the number and dry weight of root nodules at flowering (Raverkar et al. 2016). A life cycle impact assessment for the production of 1 kg of seaweed extract from the fresh biomass of *K. alvarezii* grown onshore in open sea conditions revealed that the production was environmentally sustainable, with a low carbon footprint of 118.6 kg CO_2 eq kg^{-1} dry weight (Ghosh et al. 2015).

A novel approach consisted of the selective extraction of growth factors, the method is simpler and less time-consuming than conventional solvent extraction for recovering trans-zeatin, a cytokinin from *K. alvarezii* sap, by the addition of an ionic liquid (Das et al. 2014).

Seaweed fertilizers improve morphological characteristics of the plant and yields and quality and also acts as both biostimulant and biofungicide (Sarkar et al. 2018). In medicinal plants it can affect the composition of the essential oils, i.e. in rosemary oil the percentage of β-pinene, α-phellandrene,

Table 1 Some recent examples of the application of seaweeds as fertilizers and benefits

Seaweed	Plant (beneficial effects)	References
Ascophyllum nodosum	*Triticum aestivum* (Y)	Stamatiadis et al. (2014)
Ascophyllum nodosum	*Mentha piperita*, *Ocimum basilicum* (MPh, OQ)	Elansary et al. (2016)
Ascophyllum nodosum extract and nanosize calcite	*Vitis vinifera* (PC, Ge)	Sabir (2015)
Ecklonia maxima (eckol)	*Brassica oleracea* (MPh, AF–AI)	Rengasamy et al. (2016a, b)
Gracilaria +RDF	*Vigna mungo* (Y)	Mahajan et al. (2016)
Hypnea valentiae	*Dolichos biflorus* (G, BC)	Renakabai et al. (2013)
Kappaphycus alvarezii, Gracilaria edulis	*Oryza sativa* (PR, MPh. G, Ge, NV, PR, Y)	Singh et al. (2015), Layek et al. (2017)
Sargassum johnstonii	*Lycopersicon esculentum* (PR, BC, NV)	Kumari et al. (2013)
Sargassum myriocystum	*Sesamum indicum* (Ge, Q, MPh)	Sujatha et al. (2013)
Sargassum tenerrimum	*Dolichos biflorus* (G, Y)	Renuka et al. (2013)
Sargassum tenerrimum	*Solanum lycopersicum* (G, Y, Q)	Sasikala et al. (2016)
Sargassum polyphyllum, Turbinaria ornata, Gelidiopsis sp., *Padina tetrastomatica and Gracilaria corticata*	*Vigna radiata* (G; AP, MPh)	Sarkar et al. (2018)
Seaweed extract	*Citrus sinensis* tree (Y, Q)	El-Shamma et al. (2013)
Seaweed extract	*Mangifera indica* (MPh, BC, NV)	Faissal et al. (2013)
Seaweed extract	*Malus domestica* tree (MPh)	Grzyba et al. (2013)
Seaweed extract	*Rosmarinus officinalis* (Y, OQ)	Jawaharlal et al. (2013)
Seaweed extract	*Persea americana* (FR, FW)	Morales-Payan and Candelas (2014)
Seaweed extract	*Dactylis glomerata*, *Festulolium braunii* (NV)	Ciepiela et al. (2016)
Seaweed extract	*Beta vulgaris* (Y, Q)	Ahmad et al. (2017)
Seaweed extract	*Vitis vinifera* leaf (FR, Q, AF–AI)	Calzarano et al. (2017)
Seaweed extract	*Glycine max* (PR)	Guerreiro et al. (2017)
Seaweed extracts and licorice roots	*Allium cepa* (FlR)	Babilie et al. (2015)
Seaweed extract and organic palm leaf or rice residuals	*Brassica oleracea* (G, Y, Q)	Manea and Abbas (2018)
Ulva fasciata, Sargassum wightii and *Padina boergesenii*	*Vigna* sp., *Brassica* sp., *Oryza sativa* (Y, MPh)	Muthezhilan et al. (2014)

SLF seaweed liquid fertilizer, *RDF* recommended dose of fertilizer, *AP* Activity against pathogens, *AF–AI* antifungal or antiinsecticidal, *BC* Biochemical characteristics, *FlR* flower rate, *FR* Fruit retention, *FW* fruit weight, *Ge* germination, *G* growth, *MPh* morphophysiological parameters (height, leaves number and area, root length), *NV* Nutritional value, *OQ* oil quality, *Q* quality, *PC* polen characteristics, *PR* productivity, *Y* yield

monoterpenes, 3-methylenecycloheptene, italicene, α-bisabolol (sesquiterpenes), α-thujene where significantly enhanced (Tawfeeq et al. 2016), while in mint oil increased contents of L-menthone and L-menthol and in basil oil chavicol methyl ether, linalool and cineol, whereas potentially toxic compounds content was decreased (Elansary et al. 2016).

The biomass generated by seaweed washed up on shores either due to storm events or green, red or brown tides is a good biomass to use but presents a relative high salt content which could be lowered through washing prior to composting the material. Abundant examples of the composting application of seaweeds can be found. The physicochemical and microbiological characteristics make residual seaweed suitable for agriculture purposes (Negreanu-Pirjol et al. 2011) and may provide plant nutrients and improve soil quality in coastal agroecosystems. Using seaweed as a partial source of nitrogen for sweet corn during one growing season changed soil biological and chemical properties and production yield and quality were either similar or superior to the obtained with an organic fertilizer (Possinger and Amador 2016). Seaweed waste was also utilized for incubation of the mushroom *Cantharellus* sp., which produced dense mycelia with antioxidant properties (Zhang et al. 2013).

Composting of seaweeds from eutrophication was proposed to produce a valuable organic fertilizer that enhances plant growth and crop yield (Michalak et al. 2017). Shorelines massive drifts of *Sargassum fluitans* and *Sargassum natans* were mixed with cafeteria food waste and local wood chips and composted, resulting in a product with similar or higher quality to standard (Sembera et al. 2018). Also wastes, such as those from *U. pinnatifida*, composting products via inoculation with *Bacillus* sp., *Gracilibacillus* sp. or *Halomonas* sp. has been safely reused as fertilizer (Tang et al. 2009). Seaweed composting with fish waste, and pine bark (1:1:3, volume) can be used as an organic amendment and/or growth substrate for use in ecological agriculture, based on physico-chemical, hygienic, and phytotoxic properties (Illera-Vives et al. 2013). In addition, incorporation of seaweed benefited the hydrological properties, water holding capacity, water conductivity, air porosity, and wettability by improving the hydrophobicity and reducing the contact angle when combined with hazelnut husk in compost (Ozdemir et al. 2015). In a study with *Saccharina latissima*, the use of seaweed biomass as fertilizer had the lowest environmental impact in terms of marine (Seghetta et al. 2016a, b, c).

Algal blooms, beach wrack and some algal wastes that are destined for producing fertilizers, are affected by the temporal variability in the concentrations of certain nutrients. The concentrations of toxic metals must be evaluated, and the sustainability of beach wrack exploitation must be considered (Villares et al. 2016).

3.3 Feed

For several centuries brown seaweeds washed ashore have traditionally been used as feed for farm animals. Benefits for the animals are derived from its content in minerals, trace elements, insoluble and soluble fiber and vitamins which are essential nutrients for animal growth. Use of these brown seaweeds requires a simple preparation process, consisting of washing, milling and drying before storage (Kraan and Guiry 2006). Different studies (Kraan and Guiry 2006; Ferrero et al. 2015) have evaluated the utilization of seaweed and seaweed wastes for feeding purposes. Trials that incorporated seaweed meal to different animal diets showed different benefits including an increase in the iodine content of the eggs when incorporated into poultry diets, increases in milk production in dairy cows and addition of seaweed maintained sheep weight during the winter season, increasing lambs' birth weights and wool production (Holdt and Kraan 2011).

3.3.1 Poultry

Some authors (Asar 1972) demonstrated that a 4.0% seaweed inclusion increased body weight gain; Gu et al. (1988) concluded that 2.0% of seaweed meal improved broiler performance and dressing percentage while Ventura et al. (1994) studied the effect of inclusion of *U. rigida* at 0.0, 10.0, 20.0 and 30.0% on chicken performance. It was reported that *U. rigida* decreased FI and body weight gain (BWG) and they concluded that it is negative to be included in the diet at level higher than 10.0%. Abudabos et al. (2013) evaluated the effect of substituting 1.0 and 3.0% of corn with seaweed (*U. lactuca*) on performance, carcass characteristics etc., and concluded corn meal can be replaced with *Ulva* and would improve weight gain and performance. Michalak et al. (2011) used enriched microelements to replace and investigate individual microelement ions in layer feed from *Enteromorpha prolifera* and *Cladophora*. This resulted in heavier birds, heavier eggs and improved yolk color suggesting several benefits for layer production can be gained by using Ulva species. Aguilar-Briseño et al. (2015) and Ibraheem et al. (2012) reported that Ulva (*Ulva clathrata, Ulva lactuca*) and *Caulerpa racemosa* exhibited an antiviral effect on Newcastle disease virus (NDV). In addition, *Ulva* had significant inhibitory effects on bacteria and fungi in both studies, highlighting an opportunity for use of this alga in poultry for anti-viral and gut health applications.

Yan et al. (2011) reported improvements in body weight and mortality of broilers infected with *Salmonella enteritidis* when birds were feed marine brown seaweed sodium alginate oligosaccharides prepared with purified alginate lyase in the form of the sodium salt at the rate of 0.04 and 0.2% of the diet. The positive results from this work included a decrease in caecal *Salmonella* spp. and significant increases in *Salmonella* spp. specific antibodies 10 days after treatment of both rates of sodium alginate inclusion.

A number of feeding trials confirmed the beneficial effect of seaweed byproducts as a novel animal feed supplement. When *Undaria pinnatifida* and *Hizikia fusiformis* wastes fermented with *Bacillus subtilis* and *Aspergillus oryzae* were supplemented to broilers, the body weight gain, gain:feed and immune response of seaweed by-product fed animals were higher, whereas mortality rate was lower compared to control group (Choi et al. 2014). Abbaspoour et al. (2015) showed increased egg production and quality with using red seaweeds in the diet (increased follicle production, reduced cholesterol and increased triglycerides). Changes were attributed to hypertriglyceridemia and hypocholesterolemia during biochemical transformation in laying quail. The feed supplementation with *Gracilaria* wastes on carcass characteristics and production efficiency of ducks was confirmed, lowering the fat content and raising the meat antioxidant status when supplemented at 12.5–15%, lowered costs and increased income compared with the basal diet (Santoso et al. 2016). A reduction was reported in *Escherichia coli* in caecal microflora samples, which was associated with increased lactic acid bacteria when brown seaweed polyammurate was fed to broilers (Zhu et al. 2015). Antibacterial properties in laying hens, using *Chondrus crispus* and *Sarcodiotheca gaudichaudii* to reduce *Bifidobacterium* spp. and *Clostridium perfringens*, while supporting the growth of gram positive bacteria *Bifidobacterium longum* and *Streptococcus salivariu* have been demonstrated by Kulshreshtha et al. (2014). Supplementing laying hens resulted in improved yolk weight and FCR.

3.3.2 Dairy and beef

Diet supplementation of breeding Hanwoo cows at 10% fermented *Undaria pinnatifida* by-product for 2 months before parturition until weaning of their calves resulted in greater weaning weight and average daily gain. Moreover, a serum and colostrum immunoglobulin G level in the first parity and elevation of moisture, crude fat and crude protein content of colostrum was observed (Islam et al. 2016).

Hong et al. (2015) studied the effect of supplementing with brown seaweed by-products up to 4% of basal diet in Holstein dairy cows and evaluated in in vitro batch culture rumen fermentation. The pH tended to be higher for the higher level of supplementation, whereas the concentration of ammonia nitrogen was lower compared with the control while the volatile fatty acid concentration was hardy affected. Dry matter intake, daily gain and feed efficiency during transition were not affected. The concentration of plasma progesterone levels in 4% seaweed by-product treatment increased to 158% compared with the initial level of the study. Triiodothyronine and thyroxine levels were also higher and seaweed supplementation did not affect milk yield and composition (Hong et al. 2015).

The effect of 2% *Undaria pinnatifida* by-product supplemented diet significantly improved the average daily gain and gain:feed ratio as well as serum immunoglobulin G concentration in Hanwoo steers. Chemical composition, quality of meat and carcass yield were unaffected and meat cholesterol, the myristic acid and palmitoleic acid concentration were reduced, whereas the concentration of stearic acid and linolenic acid increased (Hwang et al. 2014).

Bendary et al. (2013) reported that seaweed treatment (Kelp and Fucus mix) showed significantly better digestibility coefficients of DM, OM, CP, EE and NFE and subsequent nutritive values than premix treatment, while the control treatment showed the lowest digestibility. CF digestibility was significantly higher for seaweed treatment than that of other treatments. Seaweed treatment showed significantly the highest average daily intake of TDN and DE followed by the premix treatment, while the lowest intake was in control treatment. The highest ruminal pH values were detected with premix treatment followed by seaweed treatment, while the lowest values were observed with control treatment. Seaweed treatment recorded the highest ruminal TVFA's concentration and the lowest NH_3-N concentration followed by premix treatment, while the control had the opposite concentrations. Seaweed treatment revealed significantly the highest total protein and globulin concentrations followed by premix treatment, while the control treatment had the lowest concentration. Feeding treatments not significantly affected the concentrations of albumin, creatinine and bilirubin and the activities of AST and ALT in serum. Average daily actual milk yield and the percentages of fat, lactose, SNF, TS and ash were significantly higher and somatic cell count in milk was significantly lower for premix and seaweed treatments compared to the control treatment. Seaweed treatment recorded the highest FCM yield and protein percentage at 4% followed by premix treatment, while the control treatment had the lowest values. The amounts of DM and DCP required for producing 1 kg 4% fat corrected milk (FCM) were significantly lower for premix and seaweed treatments than those of control treatment. While, the amount of TDN and DE required for producing 1 kg 4% FCM were nearly similar for the different treatments. Feed cost (LE/day) were nearly similar for the different treatments. While feed cost per one kg 4% FCM was significantly lower, but the total and net revenue were significantly higher for premix and seaweed treatments compared to control treatment.

3.3.3 Swine

Alginate oligosaccharides, generated by the depolymerization of alginic acid by alginate lyases can be proposed as a supplement in swine feed to increase the average daily body weight gain and promote antioxidant defense properties, by enhancing endogenous enzymes (Wan et al. 2017). Several non-starch polysaccharides (NSP such as alginates, laminarin and fucoidan) in seaweeds act as bioactive molecules with a pronounced anti-microbial action, similar to in-feed antibiotics in piglets. This is beneficial from a performance perspective, as a lower microbial load will result in a lower energy cost to the pig. Moreover, the removal of harmful bacteria like *E. coli* helps control disease rates in piglets (Reilly et al. 2008; O'Doherty et al. 2010). Dierick et al. (2009) reported an improvement of pig gut health and increase of iodine in meat using seaweed while O'Sullivan et al. (2010) demonstrated antibacterial effect and prebiotic effect using brown seaweed extracts. Positive effects against scouring and diarrhea and reduced ammonia output were shown by Williams and Edyvean (1997) and O'Reilly et al. (2008).

Strong anti-helminth working against parasitic and intestinal worms was shown by Higa and Kuniyoshi (2000). O'Doherty et al. (2010) showed that the inclusion of a laminarin and fucoidan extract only increased both daily gain (+ 11%) and feed efficiency in post weaned pigs. Similar responses in growth rate and feed efficiency have been reported by McDonnell et al. (2010) with laminarin. This improved performance may be attributable to a number of reasons. First, there is a reduction in *E. coli* numbers in the gut of the laminarin and fucoidan extract-fed pigs. The inclusion of the seaweed extract decreased the counts of *E. coli* in the faces compared with control diets. Similarly, McDonnell et al. (2010) demonstrated that the inclusion of a laminarin extract had an inhibitory effect on the counts of *E. coli* in weaner pigs. The decrease in *E. coli* numbers as a result of laminarin and fucoidan inclusion was particularly evident in pig health between 7 and 14 days post weaning. The results of O'Doherty et al. (2010) showed, in pigs, a reduction in the number of *E. coli* and an increase in *Lactobacilli* spp. with dietary inclusion of the brown seaweed *Laminaria* spp. The authors reported that this was due to reduced adherence and translocation of bacteria across the epithelial wall, supporting the notion that this unique polysaccharide might offer support for improved health in animals.

3.4 Nutraceuticals, cosmeceuticals and pharmaceuticals

Seaweeds have developed efficient metabolic mechanisms to survive in an environment exposed to extreme salinity, osmotic changes during low tide, freshwater exposure, irradiation conditions and herbivory and some of these compounds, exclusive to seaweeds, are a promising feedstock for the production of novel bioactives. These bioactives include different families of compounds useful for the formulation of functional foods, cosmetics and pharmaceuticals (Charoensiddhi et al. 2017; Gade et al. 2013).

The best known example is sulfated polysaccharides from seaweeds. These bioactive molecules can act as antithrombotic (Colliec et al. 1991), antiinflammatory (Flórez-Fernández et al. 2017a, b, c; Phull and Kim 2017), antiallergic (Vo et al. 2015), anticancer (Park and Pezzuto 2013; Kwak 2014; Atashrazm et al. 2015), antioxidant (Li and Kim 2011; Balboa et al. 2013a, b), osteogenic (Chaves Filho et al. 2018) and cardioprotective agents (Mayakrishnan et al. 2013). Another polysaccharide is alginate, particularly the oligomeric fractions, has notably diverse pharmacological activities. The guluronate oligosaccharides prepared by oxidative degradation from alginate showed in vitro anti-inflammatory activity (Zhou et al. 2015). Sodium alginate was also used to prepare nanoparticles formulated with ampicillin for osteoporosis treatments (Qu 2017). The oligosaccharide has been shown to have anti-bacterial (Pritchard et al. 2017) and anti-biofilm properties and potentiates the activity of selected antibiotics against multi-drug resistant bacteria (Tøndervik et al. 2014).

Laminarin is found in the fronds of all members of the Laminariaceae and, to a lesser extent, in the Fucaceae. Laminaran does not gel but form viscous solutions and its main potential is in medical and pharmaceutical uses. Especially the use of laminarin as substratum for prebiotic bacteria seems to have a good commercial application (Deville et al. 2004). Laminarin may have value as a tumor-inhibiting agent and, in the form of a sulphate ester and as an anti-coagulant (Miao et al. 1999). Laminarin only shows anti-coagulant activity after structural modifications such as sulphation, reduction or oxidation. The anti-coagulant activity is improved chemically by increasing the degree of sulphation (Shanmugam and Mody 2000). Laminarin provides protection against infection by bacterial pathogens, and protection against severe irradiation, it boosts the immune system by increasing the B cells and helper T cells, lowers the levels of total cholesterol, free cholesterol, triglyceride and phospholipid in the liver (Miao et al. 1999; Renn et al. 1994a, b) and reduces cholesterol levels in serum and lowers systolic blood pressure, among other effects (Hoffman et al. 1995). The hypocholesterolemic and hypolipidemic responses are noted to be due to reduced cholesterol absorption in the gut (Kiriyama et al. 1969; Lamela et al. 1989; Panlasigui et al. 2003). This is often coupled with an increase in the faecal cholesterol content and a hypoglycemic response (Dumelod et al. 1999; Ito and Tsuchida 1972; Nishide et al. 1993). Laminarin as a potential cancer therapeutic is under intensive investigation (Miao et al. 1999) and demonstrates the excellent inhibitory effects on the adhesion of the ulcer-causing pathogen *H. pylori* to gastric cells. Supplementation to piglets diet has antimicrobial and anti-inflammatory activity when supplemented maternally or directly (Bouwhuis et al. 2017).

Another important group is seaweed fatty acids. These fatty acids are known to have a strong antiinflammatory working (McCauley et al. 2015) and are one of the key components of importance in neuroprotection (Cornish et al. 2017).

Brown seaweed polyphenols possess a variety of bioactivities as antioxidant and anti-inflammation effects (Balboa et al. 2013a, b; Wijesinghe et al. 2013; Casas et al. 2016), antitumoral (Pádua et al. 2015), protection against cardiovascular diseases and diabetes (Murray et al. 2018). Fucoxanthin, a carotenoid from brown seaweed shows biological activities such as antioxidant, antitumoral, anti-inflammatory and antiobesity (Pádua et al. 2015; Maeda et al. 2006; Miyashita et al. 2010; Kim and Pangestuti 2011; Gammone and D'Orazio 2015). Phycobiliproteins are oligomeric colored proteins from red seaweeds, which are extensively commercialized for natural colorants in food and cosmetics as antimicrobial, antioxidant, anti-inflammatory, neuroprotective or hepatoprotective agents and fluorescent applications in clinical and immunological analysis (Chandra et al. 2017; Sekar and Chandramohan 2008) Green and red seaweeds are an important source of proteins (Tamayo Tenorio et al. 2018). Enzyme hydrolysis of protein by-products with commercial proteases is the most usual strategy to obtain bioactive peptides, showing antioxidant, antimicrobial, antiinflammatory, antihypertensive, anticancer, and hypoglycemic action (García et al. 2016; Admassu et al. 2018), with advantages over synthetic drugs because they have low toxicity, less side effects and are more bioavailable (García et al. 2016; Pangestuti and Kim 2017).

Crude extracts showing and enhanced oxidative stability of cosmetic formulations, such as acidic extract from *Ulva lactuca* from greentides (Balboa et al. 2014), are effective in the formulation of nanoparticles, i.e., silver chloride nanoparticles with antibacterial (Dhas et al. 2014; Govindaraju et al. 2015), antidiabetic (Dhas et al. 2016), and insecticidal (Murugan et al. 2018) activities. Seaweed extracts and ingredients can have an effect on skin elasticity. Seaweed polysaccharides are recognized for their biofunctional and physicochemical characteristics (Shanura Fernando et al. 2018). The enzyme mainly responsible for collagen breakdown in skin is MMP-1 (fibroblast collagenase). Fucoidan has been shown to inhibit UVB-induced MMP-1 expression in human skin fibroblasts (Moon et al. 2008, 2009a, b). Furthermore, *Eisenia bicyclis*, *Ecklonia cava*, and *Ecklonia stolonifera* also possess strong abilities to inhibit MMP-1 expression (Joe et al. 2006).

In recent years, seaweeds have attracted great attention in search of natural tyrosinase inhibitor agents for skin whitening through melanocyte loss, and tyrosinase inhibition (Kim et al. 2014; Chang and Teo 2016). Several phlorotannins (Yoon et al. 2012) and low molecular weight fucoidans (Park and Choi 2017), have shown to be effective tyrosinase inhibitors.

Photoprotective effects through microsporyne amino acids (MAAs), carotenoids, and polyphenols have been identified from different classes of seaweeds (Ryu et al. 2009; De La Coba et al. 2009). Phlorotannins can protect cells from UVB-induced mutation and prevent chronic inflammation (Hwang et al. 2006). In Jeju Island (Korea), the leaves of *E. cava* have traditionally been used to heal sunburned skin inflammation caused by overexposure to UV (Hwang 2010). *Ecklonia cava* phlorotannins and extracts provide a strong protective effect against UVB radiation-induced cell damage in HaCaT cells and strong protective properties against photooxidative stress reduced by UVB radiation in human fibroblast cells. Furthermore, diphlorethohydroxycarmalol, a carmalol derivative from *Ishige okamurae*, demonstrated strong protective properties against UVB radiation via damaged DNA tail length and morphological changes in fibroblasts (Heo et al. 2010). Fucoxanthin and astaxanthin, two carotenoids isolated from brown seaweeds, have been demonstrate to possess photoprotective properties in human fibroblast cells via inhibition of DNA damage and enhanced antioxidant activity (Heo and Jeon 2009).

The most important step for preparing extracts and products containing bioactives is the extraction step. The complex structure of seaweed cell walls represents an important challenge for the effective extraction of these bioactives. Conventional extraction, frequently proposed has many disadvantages due to the long periods of high temperatures or harsh extractants that could cause irreversible damage, particularly to thermosensitive components, such as carotenoids or phenolics (Balboa et al. 2016; Plouguerné et al. 2008; Balboa et al. 2013a, b; Billakanti et al. 2013). Over the last 5 years, extensive research has been undertaken on the efficient extraction of seaweed bioactives using novel greener extraction processes. This includes the use of enzymes; (Hardouin et al. 2014a, b; Kerton et al. 2013; Kadam et al. 2013, 2015; Hahn et al. 2012; Díaz-Reinoso et al. 2017; Garcia-Vaquero et al. 2017), including the use of enzymes; (Hardouin et al. 2014a, b, 2016; Charoensiddhi et al. 2017; Wijesinghe and Jeon 2012; Fayad et al. 2017), fermentation (Wijesinghe et al. 2013; Mun et al. 2017), microwave assisted technology (Rodriguez-Jasso et al. 2011; Pérez et al. 2014; Lorbeer et al. 2015; Yuan and Macquarrie 2015a, b; Chakraborty et al. 2017; Magnusson et al. 2017), ultrasonic assisted processes (Fidelis et al. 2014; Topuz et al. 2016; Flórez-Fernández et al. 2017a, b, c; Mittal et al. 2017; Navya and Khora 2017), as well as green solvents and compressed fluids conditions (Balboa et al. 2013a, b, 2015; Fayad et al. 2017; Herrero and Ibáñez 2015; Klejdus et al. 2014; Conde et al. 2015), and high hydrostatic pressure (Rodrigues et al. 2017) or aqueous solutions of surface-active ionic liquids and anionic surfactants for the carotenoids extraction (Vieira et al. 2018). Table 2 summarizes some examples on the extraction of seaweed bioactives.

3.5 Biocomposites

The production of biodegradable bio-based films from renewable agro-industrial by-products and marine residues, including seaweed, can add value to what is normally considered as waste product. The packaging industry is increasingly demanding these bio-based films made from traditional waste proucts. Composites prepared with seaweeds derived polysaccharides (alginate, carrageenan, and agar) offer advantages due to their, renewability, environmental friendliness and sustainability for food packaging and for pharmaceutical uses. Examples are tissue engineering, drug delivery, and wound dressing. Seaweed polysaccharides have advantages in relation to biocompatibility, availability, gelling capacity, and encapsulation efficiency (Abdul Khalil et al. 2017; Leceta et al. 2014). The manufacture stage should be carefully optimized due to its high environmental impact (Leceta et al. 2014). The by-product from the filtration step of the agar extraction process from the red alga *Hydropuntia cornea* has been explored as inexpensive and effective filler for incorporation by melt blending into a poly(lactic acid) matrix (Madera-Santana et al. 2015). Seaweed waste from *K. alvarezii* and *G. verrucosa*, remaining from an aquaculture pond's harvest was repulped, washed, disintegrated and stirred to obtain pure seaweed fiber, which was mixed with epoxy adhesive and sawdust to produce medium density fiberboard by dry process (170 °C, 45 Pa bar, 25 min) (Alamsjah et al. 2017).

The *Posidonia oceanica* wastes, accumulated in beaches, were used at 5–40 wt%, as particle filler for

Table 2 Some examples dealing on the extraction of seaweed bioactives

Seaweed	Extraction technology	Biological properties	References
Polysaccharides			
Ascophyllum nodosum	MWAE	AO	Yuan and Macquarrie (2015a, b)
Chnoospora minima	EAE	AW, SW	Shanura Fernando et al. (2018)
Ecklonia cava	EAE, MWAE	AI	Lorbeer et al. (2015), Lee et al. (2012)
Furcellaria lumbricalis	CSE	IM	Yang et al. (2011)
Gracilaria birdiae	CSE, EAE, USAE	AC, AO	Fidelis et al. (2014)
Laminaria japonica	CSE	AI, AO	Cui et al. (2010), Zhao et al. (2018)
Lobophora variegata	EAE	AI, AT	De Sousa et al. (2017)
Saccharina japonica	SWE	AO	Saravana et al. (2018)
Sargassum muticum	AH, USAE	AO, AT	Flórez-Fernández et al. (2017a, b, c), Balboa et al. (2013a, b)
Sargassum filipendula	EAE	AM, IM	Telles et al. (2018)
Sargassum polycystum	EAE	AW, SW	Shanura Fernando et al. (2018), Alanisamy et al. (2017)
Solieria chordalis	MWAE	AH	Boulho et al. (2017)
Turbinaria decurrens	CSE	NP	Meenakshi et al. (2016)
Ulva armoricana	EAE	AO, AV	Hardouin et al. (2016)
Ulva intestinalis	USAE	AO	Rahimi et al. (2016)
Ulva lactuca	CSE, EAE,	AO	Yaich et al. (2017)
Undaria pinnatifida	MWAE		Quitain (2013)
Lipids/Carotenoids			
Palmaria palmata	CSE	AI	Robertson et al. (2015)
Porphyra dioica	CSE	AI	Robertson et al. (2015)
Sargassum muticum	CSE, IL, sc-CO_2	AO, AOb	Conde et al. (2015), Vieira et al. (2018)
Sargassum siliquastrum	CSE	AO; UVP	Heo and Jeon (2009), Heo et al. (2008)
Undaria pinnatifida	CSE; EAE	AOb	Maeda et al. (2006), Billakanti et al. (2013)
Phlorotannins			
Ascophyllum nodosum	CSE	AI, AO, AOb	Dutot et al. (2012), Austin et al. (2018)
Carpophyllum flexuosum	MWAE	AO	Zhang et al. (2018)
Ecklonia cava	CSE	AI	Wijesinghe et al. (2013), Kim et al. (2014)
Ecklonia kurome	CSE	AI, AOb	Mori et al. (2014)
Fucus spiralis	CSE	ACEI	Paiva et al. (2016)
Ishige foliacea	CSE	NP	Um et al. (2018)
Laurencia obtusa	USAE	AO	Topuz et al. (2016)
Lessonia nigrescens	EAE	ACEI	Olivares-Molina and Fernández (2016)
Sargassum muticum	AH, EAE	AO, AE, AI, AT	Casas et al. (2016), Puspita et al. (2017)
Sargassum vestitum	MWAE	AO	Ang et al. (2017)
Peptides and proteins			
Gracilariopsis lemaneiformis	EAE	ACI	Cao et al. (2017)
Grateloupia turuturu	EAE; USAE		Le Guillard et al. (2015)
Palmaria palmata	EAE	ACEI	Hamedy and FitzGerald (2013), Fitzgerald et al. (2014), Furuta et al. (2016)
Porphyra yezoensis	EAE	ACEI	Qu (2010)

Table 2 continued

Seaweed	Extraction technology	Biological properties	References
Pyropia columbina	CSE, EAE	ACEI, AO	Ian et al. (2015)
Undaria pinnatifida	EAE, CSE	ACEI	Sato et al. (2002), Suetsuna et al. (2004)

CSE conventional solvent extraction, *EAE* enzyme assisted extraction or modification, *IL* ionic liquids, *MWAE* microwave assisted extraction, *Sc-CO*$_2$ supercritical carbon dioxide, *SWE* subcritical water extraction, *PHWE* pressurized hot water extraction, *AH* autohydrolysis, *USAE* ultrasound assisted extraction, *AA* antiarthritic, *AC* anticoagulant, *ACEI* angiotensin L-converting enzyme inhibitor, *AE* antielastase, *AO* antioxidant, *AOb* antiobesity, *AH* antiherpetic, *AM* antimicrobial, *AT* antitumoral, *AV* antiviral, *AW* antiwrinkle, *IM* immunomodulatory, *NP* neuroprotective, *SW* skin whitening, *UVP* UV protection

natural fiber reinforced plastics in combination with a biobased polyethylene obtained from sugar cane as matrix. The biobased composite material could replace wood and wood like products, since it presents interesting mechanical performance and stability for outdoor applications (Ferrero et al. 2015).

Eucheuma cottonii wastes have better thermal stability, higher crude fiber content, lower moisture content and similar density to the raw seaweed, and these biomass wastes have good potential as renewable filler material, incorporated up to a 40% in a thermoplastic sugar palm starch/agar blend prepared by melt-mixing and hot pressing (140 °C, 10 min). Significant improvement in the tensile, flexural, and impact properties of the composites was observed. The addition of seaweed enhanced the thermal stability and soil biodegradation of the composites, a property of interest for short-life applications e.g., in the fast food industry such as trays, plates, etc. (Jumaidin et al. 2017a, b).

3.6 Biofuel

The methods proposed for the production of biofuels have been usually classified in biochemical and thermochemical; the first include anaerobic digestion to produce methane and hydrogen; fermentation to produce ethanol, acetone and butanol; and extraction of hydrocarbons to produce biodiesel; the second group includes combustion, liquefaction, gasification and pyrolysis (Chen et al. 2015; Michalak 2018; Sudhakar et al. 2018), but the different chemical composition compared to land biomass requires a different technological approach (Michalak 2018).

Seaweed industries aimed at obtaining food ingredients generate important amounts of waste by-products, which could present a potential source for biofuel production under the biorefinery approach. Other attractive materials are sustainable low cost sources, such as the invasive species *Caulerpa* spp. and *Sargassum* spp. and those occurring in persistent and seasonal massive blooms, such as *Ulva* sp. (Kraan and Guiry 2006). The utilization of waste material or biomass derived from alga blooms is desirable from both an economic and environmental point of view.

Most seaweeds are generally more productive than other land crops. Seaweeds can be produced in salt water with nutrients from seawater and can be adapted to live in a variety of environmental conditions. Seaweeds are regarded as a third generation feedstock for biofuels (biogas and biodiesel) from them (Kraan 2013; Goh and Lee 2010; Fernand et al. 2017; Alaswad et al. 2015), and seaweed wastes have been named “fourth generation” or “marine second generation” biofuels. The major barriers to the competitive application of seaweed for energy production are the high cost associated to seaweed farming and harvesting, the regular supply of seaweed biomass, the selection of an appropriate conversion technology for transformation in the vicinity of the origin (Goh and Lee 2010) and limitations in relation to the higher moisture and ash content of seaweeds compared to lignocellulosic plants (Kraan 2011; Kumar et al. 2013; Marquez et al. 2015). According to economic considerations and the principles of green design, the successful transformation of seaweeds to fuels needs a simultaneous production of value-added co-products (Michalak 2018; Alaswad et al. 2015; Sambusiti et al. 2015; Kumar et al. 2018) and when the biomass production increases in scale, new products for medium- and high-volume markets will be needed (Foley et al. 2011). In addition, the valorization of waste fractions from biofuel can be destined for production for environmental and wastewater bioremediation (Zeraatkar et al. 2016).

3.6.1 Ethanol and butanol

Seaweeds are good candidates for the conversion of their polysaccharides into sugars which can then be converted into ethanol (Goh and Lee 2010). However, the variety of carbohydrates makes it difficult for conversion into simple sugars and coupled to this the choice of appropriate microorganisms (Jung et al. 2013). Seaweeds are economically more attractive than woody lignified biomass (Baghel et al. 2016), and the use of waste materials is favored (Seghetta et al. 2014; Kang and Kim 2015). The conversion of biomass to bioethanol usually requires different stages: (1) pretreatment of feedstock to release complex carbohydrates; (2) hydrolysis of polysaccharides to monosaccharides; (3) fermentation and (4) product recovery (Harun et al. 2014).

Pretreatment The application of similar procedures as those described for saccharification of lignocellulosic biomass was tried with seaweeds, i.e. hydrothermal pretreatment, wet oxidation, steam explosion, plasma-assisted pretreatment and ball milling (Harun et al. 2014; Schultz-Jensen et al. 2013). However, adequate optimization is required since important losses could occur, i.e. wet oxidation of *Chaetomorpha linum* caused more than 50% biomass loss (Schultz-Jensen et al. 2013). Acidic pretreatment at high temperature is widely used, but requires certain chemicals which are difficult to recover and can generate non-sugar byproducts, with inhibitory potential on further biological conversion. Alternatively, gamma irradiation is an effective method for the depolymerization of complex polysaccharides and structural breakage of the seaweed cell wall (Yoon et al. 2012). Other pretreatments proposed for altering enzymatic digestibility and ethanol potential from the green seaweeds are ethanol organic solvents and liquid hot water, causing more drastic structural changes than pretreatments with alkaline media and ionic liquids (Jmel et al. 2018).

Hydrolysis Dilute-acid hydrolysis is a typical physicochemical method to treat raw seaweeds biomass (Yanagisawa et al. 2011; Meinita et al. 2012a, b; Park et al. 2011; Lee et al. 2013), and is highly influenced by acid concentration and hydrolysis time which need to be optimized to maximize concentrations of monosugars and ethanol (Meinita et al. 2012a, b). Combination with ionic liquid pretreatment increased the enzymatic saccharification of seaweed waste pretreated (Uju Wijayanta et al. 2015). The decomposition of sugars caused by acid treatment at elevated temperatures can lead to the formation of degraded products, susceptible of causing inhibition of fermenting microorganisms or inducing a prolonged lag phase, i.e. furfural, 5-hydroxymethylfurfural and levulinic acid. Metals in seaweeds can also act as inhibitory substances, and show a higher content (0.5–11%wt.) than for terrestrial biomass (1–1.5%wt.) (Jung et al. 2013). Degraded products in pretreated hydrolysate can be removed prior to fermentation, and activated charcoal and calcium hydroxide have been frequently proposed (Meinita et al. 2012a, b; Ra et al. 2015, 2017), as in the detoxification of hydrolysates from lignocelullosic biomass (Soto et al. 2011). Less often used is the alkaline treatment, i.e. for galactan extraction (strong alkaline solution, 70–90 °C, 5 h), but the major disadvantages are the low polysaccharide yields and generation of waste streams (Goh and Lee 2010). The efficient hydrolysis to monosaccharides or polysaccharides without a fermentation inhibitor was reported using an integrated pretreatment with hydroxyl radicals and hot water (Gao et al. 2015). Alternatively, the production of platform-chemicals and sugar production by dilute-acid-catalyzed hydrothermal reaction was proposed. *Kappaphycus alvarezii* was used for the production of glucose, galactose, levulinic acid and 5-hydroxymethylfurfural (Lee et al. 2016a, b).

Enzymatic hydrolysis is a milder saccharification tool for both whole seaweeds or for residual fractions, but the lack of specific enzyme activities for seaweed polysaccharides limits this approach and the contribution of molecular bioengineering would be interesting since new microorganisms would be required (Jung et al. 2013; Kim et al. 2015; Shukla et al. 2016).

The hydrolytic enzymes used for lignocellulosics (cellulase and cellobiase) and other multienzyme complexes are frequently proposed for seaweeds (Yanagisawa et al. 2011; Trivedi et al. 2013). Other specific enzymes for the degradation of brown algal polysaccharides, are alginate lyases, laminarinases or β-glucanases, isolated from marine microorganisms (Jung et al. 2013; Wargacki et al. 2012; Kim et al. 2013a, b), seaweed compost (Tang et al. 2009) or sandbar (Kang and Kim 2015), and could be suitable to saccharify seaweeds although they showed low hydrolysis efficiency, requiring additional pretreatment (Jung et al. 2013; Schaumann and Weide 1990; Adams et al. 2011). The enzymes represent an important cost of the process, therefore reutilization is encouraged (Trivedi et al. 2013). Combinations of treatments can be successful, such as mechano-enzymatic (Amamou et al. 2018) or chemical and enzymatic hydrolytic treatments (Jung et al. 2013; Ra et al. 2015; Ge et al. 2011; Sunwoo et al. 2016). The absence of pretreatment prior to fermentation yielded lower ethanol values, but the direct application reduces the product cost (Kang and Kim 2015).

Fermentation to ethanol is generally performed by Saccharomyces cerevisiae, commonly used for industrial fermentation of glucose, but also can ferment galactose (Goh and Lee 2010). Seaweeds contain specific carbohydrates such as mannitol and laminaran which are readily utilized (Horn et al. 2000; Lee and Lee 2012), and in some cases, previous adaptation of the strains to special sugars and to the high salinity was required (Cho et al. 2013; Sunwoo et al. 2017). Furthermore, the utilization of seaweed mixtures and different microorganisms has been reported, i.e. a mixture of red, brown, and green seaweed wastes treated with acid, saccharified with enzymes and further fermented with a co-culture of adapted *S. cerevisiae* and *Pichia angophorae* (Sunwoo et al. 2017). A novel approach consists of the use of metabolically modified bacteria (Takeda et al. 2011). Conditions for hydrolysis and fermentation in studies published from 2000 to 2015 (Fernand et al. 2017) are summarized in Table 3.

The bioconversion strategies proposed are separate hydrolysis and fermentation (SHF) and simultaneous saccharification and fermentation (SSF). Furthermore, consolidated bioprocessing, consisting on performing enzyme production, hydrolysis, and fermentation in a single unit, has been proposed (Fernand et al. 2017; Harun et al. 2014). Most processes were successfully scaled up (Shukla et al. 2016). After the biomass is converted to ethanol, the solid protein residue can be applied as animal feed in particular for fish feed (Seghetta et al. 2016a, b, c).

Through the acetone–butanol–ethanol process, seaweeds can act as a substrate for biobutanol production, a bulk chemical and biofuel. Identification of microorganisms that can metabolize seaweed carbohydrates is of great importance as some anaerobic bacteria such as *Clostridium* sp. do not effectively utilize some glucose-based polysaccharides, such as mannitol (Jung et al. 2013; Huesemann et al. 2012). The toxicity of butanol to microorganisms poses another problem; therefore, integration of strategies for product recovery, such as adsorption, gas stripping, liquid–liquid extraction, and pre-vaporization have been proposed. Metabolic engineering tools have aided in the development of strains with improved performance on fermentation of these substrates (Lee et al. 2016a, b). Similar pretreatment conditions as those selected for bioethanol production, i.e. thermal acid have been reported (Dubey et al. 2015). Regarding the bioconversion requirements, *Ulva lactuca* hydrolysate were fermentable to acetone, butanol and ethanol without nutrient supplementation using *Clostridium* sp. Depending on the species, different final products could be obtained. The monosaccharide composition has an influence on the main fermentation product (Van der Wal et al. 2013).

Seaweeds can be the source for other metabolites in bioconversion processes, i.e. succinic acid from *S. latissima* biomass. *Saccharina latissima* also has a high phenolic content especially when harvested in summer. Post-hydrolysis of solid residue of *S. lattisima* rich in minerals, can contribute to the economic viability of the production of succinic acid and which can be optimized when mineral fertilizers and antioxidants are coproduced (Marinho et al. 2016). Seaweeds biomass is used as carbon and nitrogen source for the biotechnological production of enzymes, i.e. pectinases using submerged and solid state fermentation of different brown seaweeds (*Dictyopteris polypodioides*, *Sargassum wightii* and *Dictyopteris divaricata*) and green seaweeds (*Ulva lactuca* and *Codium tomentosum*) (Pervez et al. 2017), lipases produced by *Staphylococcus xylosus* and *Rhizopus oryzae* from *Ulva rigida* biomass and the supernatants generated from tuna by-products (Sellami et al. 2013).

3.6.2 Biodiesel

Biodiesel obtained by transesterification (alcoholysis) of triglycerides is derived commercially from animal

Table 3 Some examples of seaweeds utilized for the production of bioethanol

Seaweed or seaweed waste	Pretreatment/Hydrolysis (conversion, %)	Fermentation (ethanol yield, %)	References
Alaria crassifolia	Thermal acid/enzymatic (82)	*Saccharomyces cerevisiae* (38)	Yanagisawa et al. (2011)
Gelidia dura residue after agar extraction	Alkaline/acid treatment (0.85)	*Saccharomyces cerevisiae* (46)	Baghel et al. (2015)
Gelidium amansii	Thermal acid/enzymatic (65)	*Saccharomyces cerevisiae* (49)	Sunwoo et al. (2016)
Gelidium amansii	Thermal/acid (72)	*Pichia stipitis* (50)	Sukwong et al. (2018)
Gellidium pusillum residue after agar extraction	Enzymatic (84)	*Saccharomyces cerevisiae* (47)	Baghel et al. (2015)
Gracilaria verrucosa	Thermal acid/enzymatic (75)	*Saccharomyces cerevisiae* (48)	Sunwoo et al. (2016)
Gracilaria verrucosa residue after agar extraction	Enzymatic (87)	*Saccharomyces cerevisiae* (43)	Kumari et al. (2013)
Gelidium elegans	Thermal acid/enzymatic (67)	*Saccharomyces cerevisiae* (38)	Yanagisawa et al. (2011)
Eucheuma denticulatum	Thermal acid/enzymatic (53–80)	*Saccharomyces cerevisiae* (47)	Sunwoo et al. (2016), Ra et al. (2017)
		Scheffersomyces stipitis (48)	
Eucheuma spinosum	Thermal acid/enzymatic (49)	*Candida tropicalis* (40)	Ra et al. (2015)
Kappaphycus alvarezii	Thermal acid/enzymatic (53)	*Saccharomyces cerevisiae* (47)	Sunwoo et al. (2016)
Laminaria digitata	Acid (28)	*Saccharomyces cerevisiae* (38)	Schiener et al. (2016a, b), Kostas et al. (2017)
	Acid/enzymatic (94)	*Saccharomyces cerevisiae* (94)	
Laminaria japonica	Acid/enzymatic (37)	Saccharomyces cerevisiae (41)	Kim and Pangestuti (2011)
Laminaria hyperborea	Acid (35)	*Saccharomyces cerevisiae* (29)	Schiener et al. (2016a, b)
Chaetomorpha linum	Hydrothermal treatment (100)[a]	*Saccharomyces cerevisiae* (39)[b]	Schultz-Jensen et al. (2013)
	Wet oxidation (98)	*Saccharomyces cerevisiae* (44)	
	Steam explosion (92)	*Saccharomyces cerevisiae* (38)	
	Plasma assisted treatment (100)	*Saccharomyces cerevisiae* (41)	
	Ball milling (91)	*Saccharomyces cerevisiae* (44)	
Gelidiella acerosa residue after agar extraction	Enzymatic (83)	*Saccharomyces cerevisiae* (47)	Baghel et al. (2015)
Kappaphycus alvarezii	Thermal acid (31)	*Saccharomyces cerevisiae* (40)	Khambhaty et al. (2012)
Saccharina japonica	Thermal acid (21)	SSF—*Bacillus* sp. + *Pichia angophorae* (33)	Jang et al. (2012)
	Thermal acid/*Bacillus* sp. (31)		
	Thermal acid/enzymatic (69)		
Saccharina latissima	Acid (26)	*Saccharomyces cerevisiae* (13)	Schiener et al. (2016a, b)
Ulva fasciata	Thermal + enzyme (91)	*Saccharomyces cerevisiae* (45)	Trivedi et al. (2013)
Ulva pertusa	Thermal acid/enzymatic (59)	*Saccharomyces cerevisiae* (38)	Yanagisawa et al. (2011)
Ulva rigida	Thermal/acid (64)	*Pachysolen tannophilus* (50)	El Harchi et al. (2018)

[a,b] Relative to glucan

fats and plant oils in the international market (Knothe et al. 2005). Seaweeds generally have a low fat content and are more suited for biogas and bioethanol rather than biodiesel. Although the production of biodiesel is well-developed from microalgae (Chisti 2007), different species of seaweeds have been used for this purpose as well, i.e. *Chaetomorpha antennina*, *Gracilaria corticata* (Sharmila et al. 2012), *Stoechospermum marginatum* (Hariram et al. 2018), *Caulerpa pelata* (Renita et al. 2017), *Ulva lactuca*, *Padina boryana* and *Ulva intestinalis* (Abomohra et al. 2018).

Usually the process consists of a two stage; extraction of the lipid fraction using an organic solvent and/or mixtures of solvents and a further transesterification using methanol and a catalysts (either commercial being the most common NaOH or even from low cost feedstocks, such as waste eggshells) (Renita et al. 2017). A single stage for the simultaneous oil extraction and transesterification was also proposed (Maceiras et al. 2011). A two-step process assisted by ultrasound was proposed to produce biodiesel from *Caulerpa peltata*, pretreating algal oil to reduce acid value followed by transesterification (Tamilarasan and Sahadevan 2014). The oil-free residual biomass can be readily converted into fermentable sugars or biogas (Abomohra et al. 2018), or for pellet manufacturing (Maceiras et al. 2011). An alternative approach consisted on the production of lipids with the oleaginous yeast *Cryptococcus curvatus*, as reported for *Laminaria japonica* (Xu et al. 2014). The utilization of dried biomass offers higher yields than from wet biomass and the adequate selection of the solvents and operational conditions can enhance the production yields (Chen et al. 2015).

3.6.3 Biogas

The energy conversion via anaerobic digestion is highly efficient and the biochemical composition of seaweeds is suitable for this application, and compares favorably with land biomass, attaining methane yields (0.31–0.48 m^3 kg^{-1} volatile solids, VS), that compared to land-based biomass such as grass or wood (0.32–0.42 m^3 kg^{-1} dry weight (Hughes et al. 2012; Jung et al. 2013; Charlier 1991; Costa et al. 2012; Langlois et al. 2012; Jard et al. 2013). Note here that when no indicated dry weight were the presented units. This route could be proposed when the levels of heavy metals in seaweed washed ashore on beaches are high and their use as food is not recommended (Balina et al. 2016). A study with the brown seaweed *L. japonica* demonstrated that the anaerobic digestion unit has the highest energy demand in the entire process and consumes approximately 14% of all electricity generated. Besides the seaweed was also the largest cost component in the process. Therefore, underutilized seaweeds and industrial seaweed wastes could be suitable for biogas production (Barbot et al. 2016), and not whole species used for human food. This provides an option helping in environmental restoration and climate mitigation (Jonouchi et al. 2006; Czyrnek-Delêtre et al. 2017; Seghetta et al. 2017).

The major variables influencing biogas production include the pretreatment, salt concentration, external nutrients, inoculum source, solid loading, residence time, temperature, and the operational configuration (Ghosh et al. 1981; Fasahati et al. 2017). Seaweed is very much a seasonal feedstock, so in order to provide a constant supply of seaweeds for biogas production one option is ensiling of seaweed, which will lower salt concentrations reducing energy losses in the process. As a matter of fact, the products of silage fermentation increased methane yields by up to 28% and compensated for volatile solid losses during ensiling (Herrmann et al. 2015; Milledge and Harvey 2016a, b).

The type and composition of the seaweed also influences the methane yield (Habig et al. 1984). Different biodegradability was reported for different seaweeds, i.e. *Fucus vesiculosus* and *Ascophyllum nodosum* showed lower biodegradability (< 30%) than *S. latissima* and *Alaria esculenta* (80%) (Ometto et al. 2018a, b). The average methane yields depend on the seaweed and bioconversion factors, yielding values in the range 0.10–0.28 m^3 CH_4 kg^{-1} volatile solids (VS) (see Table 4).

Other compilations are found in the literature (Chen et al. 2015). Several factors are clearly affecting methane yields, including the seasonal variation of seaweeds composition (Czyrnek-Delêtre et al. 2017; Ometto et al. 2018a, b) and the presence of some components acting as antimicrobial or toxic substances. Polyphenols and divalent metal ions affect anaerobic digestion of brown seaweeds reducing yields and showing a lag phase (Moen et al. 1997); their polymerization degree also influenced the bactericidal effect of *L. digitata* as a result affecting cell membrane permeability, causing disruption of cell membranes (Hierholtzer et al. 2013). Appropriate adaptation strategies have to be considered, i.e. starting with low proportion of seaweed was suggested when *L. digitata* and green peas were codigested (Gurung et al. 2012; Akunna and Hierholtzer 2016).

Different pretreatments have been developed to enhance the anaerobic digestion of seaweeds, including mechanical, thermal [60–220 °C: (Vivekanand et al. 2012; Adams et al. 2015)], chemical pretreatment with acids or with alkalis, enzymatic digestion (Karray et al. 2015) and fermentation treatments. Pretreatment type and time should be adapted to each seaweed considering that pre-treatment influences the technical, economic and environmental sustainability of the process. Particle size reduction can improve biogas and methane yield, but no general recommendation on the ideal particle size has been established, since the energy demand should also be minimized (Tedesco et al. 2014). Washing seaweeds is a usual pretreatment to remove salt and to improve the volatile solids content (Tabassum et al. 2017), but careful optimization is required since it removes particulate matter and may also remove water-soluble carbohydrates. For example, up to 49.3% laminarin content of *L. digitata* was washed away. Moreover, the process has to be adapted to the seasonal variations in composition as well (Adams et al. 2015). Thermal pretreatment of *S. latissima* increased the methane yield and codigestion with steam-exploded wheat straw also had a clear positive effect on biogas production in relation to the untreated substrates (Vivekanand et al. 2012). Combination of techniques was also feasible, e.g. chemo disperser liquefaction coupling a mechanical dispersion with a chemical treatment with sodium tripolyphosphate reduced the cost and specific energy demand of liquefaction (Tamilarasan et al. 2017), and mechanical and microwave pre-treatment improved yields (Romagnoli et al. 2017). A different alternative for the selective biomethanation of the hydrolysis juice produced from *Ulva biomass* could be used for methane production instead of using the entire alga (Morand and Briand 1999) Alternatively, waste acidic fractions can be used and increase methane production. Barbot et al. (2014) reported a thermo-acidic pretreatment using either 0.2 M HCl or used a low cost acid from flue gas condensate, an industrial waste from power plants, resulting in increased methane yield in biogas production than with untreated biomass. However, pretreatment was not required for some substrates, such as the industrial *L. japonica* waste (Barbot et al. 2015).

The low C:N ratio found in some seaweeds could require additional supplementation, i.e. the C:N ratio (8:1) of *S. muticum* provided low methane under 0.11 m^3 CH_4/kg VS (Milledge and Harvey 2016a, b), and different proposals of codigestion have been published e.g., *Sargassum* sp. with glycerol and waste frying oil (Oliveira et al. 2015). The wastes from the industrial alginate production yielded a conversion of 0.27 m^3 CH_4 kg^{-1} VS and required nutrient addition such as mineral solution or pig manure (Carpentier et al. 1988). Production of biomethane from monodigestion of *U. lactuca* was also problematic due to high levels of sulphur and mixed with 3:1 ratio of dairy slurry, yielding 0.17 m^3 CH_4 kg^{-1} VS (Allen et al. 2014). Co-digestion of acid catalyzed steam pretreated and enzymatic hydrolyzed wheat straw and seaweed hydrolysates maintained a stable anaerobic process (Nkemka and Murto 2013). Co digestion of *Laminaria* sp. and *Ulva* sp. with milk wastes did not result in high methane concentration but this strategy was effective in suppressing fluctuations in the supply of seaweeds (Matsui and Koike 2010). Co-digestion of *S. latissima* with mixed municipal wastewater sludge (in mesophilic and thermophilic conditions) produced an average methane yield of 0.22 and 0.25 m^3 kg^{-1} VS in mesophilic and thermophilic conditions, respectively (Ometto et al. 2018a, b). Ulva utilization for biogas production could be favored by the low amounts of phenolic components and a large saccharide content. A low methane yield (0.20 m kg^{-1} VS) and the low utilization (50% VS), could be due to the low density of alga in suspensions, to the high S concentration leading to the production of a biogas with a high H_2S conten (Briand and Morand 1997). Pig slurry was proposed as co-substrate for anaerobic digestion (52/48, wt/wt), although the produced biogas contained 3.5% H_2S, and required a treatment before utilization for energy recovery. Temporal lowering of H_2S emissions were recorded with the sequential addition of potassium molybdate as sulphate reduction inhibitor (Peu et al. 2011). Methane from *Gracilaria vermiculophylla* after physical pre-treatment (washing and maceration) reached 0.48 m^3 CH_4 kg^{-1} VS and co-digestion with 2% glycerol increased the methane production by 18%, and co-digestion with secondary sludge (15:85, wt%) increased the methane production 25% (Oliveira et al. 2014).

Most studies on the anaerobic digestion of seaweeds are performed in batch equipment, but anaerobic digestion was also performed in cascading biomethane systems (Wall et al. 2017), sequencing batch (Jung et al. 2017) and in continuous operation (Nkemka and Murto 2013; Jung et al. 2017). Anaerobic digestion of *Ulva* biomass in a sequencing batch allowed a more efficient and stable operation and higher methane productivity and biomass retention, with higher sulfide production, probably due to the higher microbial diversity and higher concentration of

Table 4 Examples of methane production from seaweeds feedstocks and pretreatments reported

Seaweeds	Pretreatment[a]	Methane yield (m^3 kg^{-1} VS)[b]	References
Ascophyllum nodosum	Es	0.18	Herrmann et al. (2015)
Ascophyllum nodosum	Mc	0.17	Montingelli et al. (2017)
Fucus vesiculosus	TA	0.12	Barbot et al. (2014)
Gracilaria vermiculophylla	W, M	0.48	Oliveira et al. (2014)
Green seaweed	–	0.35–0.48	Hansson (1983)
Laminaria digitata	D	0.33	Edward et al. (2015)
Laminaria digitata		0.36	D'Este et al. (2017)
Laminaria digitata	HW	0.28	Tabassum et al. (2017)
Laminaria digitata	–	0.22	Guneratnam et al. (2017)
Laminaria hyperborean	Mc, T	0.06	Sutherland and Varela (2014)
Laminaria japonica	U or TA	0.17	Barbot et al. (2015)
Laminaria sp.	Mc	0.24	Montingelli et al. (2017)
Macrocystis pyrifera		0.22	Fan et al. (2015)
Laminaria digitata	Es	0.34	Herrmann et al. (2015)
Laminaria saccharina	–	0.28	Jard et al. (2013)
Palmaria palmata			
Saccharina latissima	U	0.22	Vivekanand et al. (2012)
	T	0.27	
Saccharina latissima	Es	0.33	Herrmann et al. (2015)
Saccharina latissima		0.28	D'Este et al. (2017)
Saccorhiza polyschides	Es	0.36	Herrmann et al. (2015)
Sargassum muticum	Es	0.11	Milledge and Harvey (2016a, b)
Sargassum sp.		0.18[b]	Oliveira et al. (2015)
Ulva lactuca		0.24	Jard et al. (2013)
Ulva lactuca	Es	0.25	Herrmann et al. (2015)
Ulva rigida	Es	0.63	Karray et al. (2015)
Ulva sp.	–	0.2	Briand and Morand (1997)
Ulva sp.	H	0.33	Morand and Briand (1999)
Ulva sp.	Mc	0.15	Peu et al. (2011)
Wastes alginate production	–	0.27	Carpentier et al. (1988)

[a] *D* drying, *E* enzymatic, *Es* ensiling, *HW* hot water washing, *H* hydrolysis, *M* maceration, *Mc* mechanical, *U* untreated, *T* Thermal, *TA* thermoacidic, *W* washing

[b] m^3 CH_4 kg^{-1} COD

methanogenesis than in the continuous mode (Jung et al. 2017). The two-stage mesophilic fermentation of *L. digitata*, comprising a hydrolysis reactor followed by two methanogenesis reactors, provided a methane concentration 22% higher than for the single-stage system (Guneratnam et al. 2017). Improved methane productivity from brown seaweeds under high salinity in a fed-batch equipment was reported after ten cycles, the methane production rate increased 8-fold and level of salinity 1.6-fold (Miura et al. 2015). This rate was enhanced in high density biomass bioreactors, using bacteria encapsulated in spherical capsules of alginate, using chitosan or Ca^{2+} as counter-ions, together with the addition of carboxymethylcellulose. An alternative support was a hydrophilic membrane (Youngsukkasem et al. 2012).

Different inoculum types have been proposed and the seaweeds-to-inoculum ratio is of importance (Romagnoli et al. 2017). A mixed inoculum was prepared from sediments, rotting seaweed, sewage sludge and rumen contents for digestion of ground green seaweeds without any nutrient addition, and without lag stage, both in mesophilic as well as in thermophilic conditions. Under these conditions, gas yields were 0.25–0.35 m^3 CH_4 kg^{-1} VS and the volatile solid reduction was around 50–55% (Hansson 1983). Most studies are carried out at mesophyllic and thermophilic conditions but Sargassum digestion at room temperature was also feasible (Tarwadi and Chauhan 1987). Different inocula can be proposed for the anaerobic digestion of *L. hyperborea*, particularly those found in an inoculum of slurry from a human sewage anaerobic digester and the rumen from seaweed-eating sheep. An inoculum from seaweed, provided higher acetate production and methane production in a two phase system, and the presence of efficient seaweed polysaccharide digesting bacteria and methanogenic archaea offered increased methane productivity (Sutherland and Varela 2014). Marine sediments are a good source of methanogenic bacteria for anaerobic treatment of *S. japonica* under high-salt conditions, with methanogenics of the *Methanosarcina* and the *Methanosaeta* genus dominating after cultivation (Miura et al. 2014, 2015). Littoral sediment, containing hydrogenotrophic methanogens predominance over acetotrophic ones, showed higher methane production from *Macrocystis pyrifera* in seawater than sublittoral ones, with a value similar to that achieved in freshwater with desalted seaweeds (Fan et al. 2015).

An important factor is the presence of salt. Anaerobic digestion of *Sargassum* sp. was affected by the sodium concentration, with a maximum methane yield of 0.29 m^3 CH_4/kg volatile solids was obtained at a sodium concentration of 4.42 g L^{-1}, with *Methanosaeta* and *Methanosarcina* being the predominate methanogens (Zhang et al. 2017). When Marquez et al. (2013) compared the methane yield of beach cast materials (dislodged sea grasses and seaweeds) seeded with inocula from cow manure, marine sediment, and beach casted associated microflora, they found that the best microbial source was the marine sediment, however the salinity (42‰) was higher than the average seawater value (34‰) and methanogenic activity could have been inhibited. Samples were given a range of pre-treatments from washing, drying and macerating. Dried *L. digitata* with 68.14% VS exhibited the highest biomethane potential of 141 ± 5.77 L CH_4 kg^{-1} VS, with methane content rising to about 70%, whereas the lowest biomethane potential of 93.35 ± 5.03 L CH_4 kg^{-1} VS with methane content of about 65% was obtained for fresh *Laminaria digitata* with 72.03% VS (Edward et al. 2015).

Combined production of biodiesel and bioethanol or biogas were obtained from *Ulva lactuca*, *Padina boryana* and *Ulva intestinalis* with high significant lipids and fatty acid methyl esters. This was useful for production of biodiesel with international standard characteristics. The oil-free residual biomass can be converted into fermentable sugars or biogas (Abomohra et al. 2018). Seasonal variations have to be taken into account also for this application. When *L. digitata* was used as a feedstock for ethanol and for methane production, minimum yields were found when the seaweed was harvested in March, since the laminarin and mannitol contents were lowest, whereas in July the highest laminarin and mannitol content provided maximum yields (Adams et al. 2011). Mannitol and laminaran were consumed faster than alginate, which was severely depolymerized, since alginate lyase activity was found (Østgaard et al. 1993). These authors found that anaerobic digestion of *S. Lattisima* carried out in batch cultures provided double methane yield from the autumn material compared to that of the spring material, but in semi-continuous operation yields were more similar.

An alternative transformation process consisted on the production of carboxylic acids in a novel bioenergy process for anaerobic co-digestion of food industry residues (pectin and carrageenan wastes), manure and beach-cast seaweed. This concept can contribute to strengthening the linkages between climate change mitigation strategies and coastal eutrophication management in an integrated holistic planning (Kaspersen et al. 2016). Carboxylic acids are platform chemical intermediates which can be converted to alcohol fuels and chemicals. The MixAlco^{TM} process converts any biodegradable material into mixed alcohol fuels through anaerobic digestion and chemical upgrading. Carboxylic acids were produced from an alkaline-pretreatment of the waste biomass from carrageenan processing through anaerobic digestion using a mixed-culture of marine-derived microorganisms. Operating at mesophilic (35 °C) conditions gave a higher acid yield with higher percentage of longer chain organic acids, such as acetic, propionic and butyric acids (Karunarathne et al. 2011).

In a common approach to sustainability and circular economy seaweeds can be processed for biogas production and the digestate can be further used as a nutrient source in soil, based on its content on phosphorus and other nutrients (Seghetta et al. 2017). The biomethanation residue from the industrial *L. japonica* waste was useful as biofertilizer, with enriched amounts of potassium, sulfur and iron (Barbot et al. 2015). Co-digestion of *L. digitata* with bovine slurry in a batch two-phase anaerobic digestion, producing 0.22 m^3 CH_4 kg^{-1} VS yielded an effluent or digestate that was used as bio-fertilizer for ryegrass and sunflower (Vanegas and Bartlett 2015). However, the load of cadmium and arsenic, could limit the utilization of the digestate in arable lands (Vanegas and Bartlett 2015). However, the load of cadmium and arsenic, could limit the utilization of the digestate in arable lands (Ometto et al. 2018a, b). When the digestate is used for fertilization purposes, low heavy metal content is desirable. A less heavy metal loaded bio-based fertilizer can be obtained after lowering the cadmium content in biomass used for biogas production. To remove Cadmium seaweed was hydrolyzed in water or in 0.1 M HCl at room temperature during 48 h after which the hydrolysate was treated with a macroporous polymer supporting titanate nanotubes, removing 94% Cd^{2+} from seaweed hydrolysate and without significantly decreasing methane production (Önnby et al. 2015). Selective removal of Cadmium did not affect other valuable co-existing ions and this step did not decrease the bio-methane yield. The final bio-fertilizer product demonstrated a high quality. Heavy metal contaminants in seaweed can also be removed by extraction to a liquid phase followed by adsorption to improve the fertilizer quality (Nkemka and Murto 2012). Hydrolyzed seaweed, by combined leaching and autoclave/alkaline treatments can be anaerobically digested for the production of methane while using an iminodiacetic acid polyacrylamide cryogen to remove Cd^{2+}, Cd^{2+}, Cu^{2+}, Ni^{2+} and Zn^{2+} ions. The effect of removing the heavy metals from seaweed leachate was confirmed in both batch and continuous operation in an up flow anaerobic sludge blanket reactor and did not affect the methane yield (Nkemka and Murto 2010).

3.6.4 Biohydrogen

The production of biohydrogen from seaweeds through dark fermentation is a simple process that is attracting increasing interest, requires low energy and can use different types of biomass, including monosaccharides and polysaccharides, following two common pathways: one producing acetate and the second butyrate. Although the hydrogen producing bacteria in pure cultures used with pure sugar substrates offered yields up to 3.8 mol H_2 mol^{-1} glucose eq., the utilization of mixed cultures from natural environments is a more practical approach. These cultures are mainly composed by microorganisms belonging to the genus *Clostridia* and *Bacillus* and treatments to inhibit H_2 competing bacteria, such as methanogenics, from the inoculum are proposed. To overcome limitations related to the efficient conversion of polymeric carbohydrates into monomeric sugars, physical, chemical and biological pretreatments are needed and to compensate production cost (Sambusiti et al. 2015; Kumar et al. 2018; Park et al. 2011; Shobana et al. 2017). Acid and heat treatment are the most used and proved superior to alkaline or ultrasonic pretreatment (Liu and Wang 2014), enzymatic hydrolysis (Nguyen et al. 2010) and electric fields (Jeong et al. 2012). Pretreating to remove microbial inhibitors and enhance the accessibility of the microorganisms involved in the process to the substrates was suggested to increase the product yield, i.e. removal of phenolic content (Radha and Murugesan 2017) or removal of 5-hydroxymethylfurfural (Park and Pezzuto 2013).

To increase H_2 yields, efficient pretreatments for biomass and improved reactor designs are proposed. Temperature, pH, organic loading rate, chemical composition, C/N ratio, salinity, heavy metals can all significantly affect the performances and efficiency of the process (Sambusiti et al. 2015; Shi et al. 2011). Usually seaweeds are washed with fresh water, dried at room temperature or in oven and milled (Sambusiti et al. 2015). This stage limits the negative effect of salt on the process. Normally salinity influences the dark fermentation efficiency, at Na^+ concentration higher than 2 mg L^{-1}, however adaptation to salty environments allowed operation at much higher concentrations of NaCl (9–75 g NaCl L^{-1}). Among the different seaweeds assayed (*Codium fragile*, *G. amansii*, *Porphyra tennera*, *Gracilaria verrucosa*, *Hizikia fusiforme*, *Ecklonia stolonifera*, *Ulva lactuca*, *Undaria pinnatifida* and *Laminaria japonica*), *L. Japonica* was selected (Park et al. 2009; Jung et al. 2011a, b; Şentürk and Büyükgüngör 2013). Results from recent literature are summarized in Sambusiti et al. (2015) and Shobana et al. (2017) and also presented in Table 5, including the operation conditions during fermentation, where pH refers in most studies to the initial pH. The mixture of hydrogen and methane, hythane, can be a more favorable fuel than methane, since it can reduce the ignition temperature and energy and improve combustion efficiency (Guneratnam et al. 2017). Biohythane production from *Sargassum* sp. was developed in first stage of hydrogen dark fermentation by *Caldicellulosiruptor saccharolyticus* with a yield of 91.3 L kg^{-1} and the end products were used for biogas production with a yield of 0.54 m^3 kg^{-1} (Costa et al. 2015).

3.6.5 Thermo-based processes

Direct combustion is the simplest energetic application of algal biomass, but the high ash content and varying biomass composition is a problem for thermal energy conversion, and seaweeds pellets cannot be used directly in boilers (Maceiras et al. 2015). However, the ash composition of *S. latissima* was found suitable for co-combustion with *Miscanthus giganteus* in a bubbling fluidized bed reactor (Skoglund et al. 2017). In addition to direct burning of biomass, energy can be produced through different thermochemical technologies, such as pyrolysis, liquefaction, and gasification (Xu et al. 2013; Plis et al. 2015). These thermomechanical processes are also proposed for the conversion of algal wastes to solid products such as biochar and activated carbon, which can be used as energy source, adsorbent and soil improvers (De Ramon and Iese 2015). However, one important deterrent of these uses is in relation to the high energy consumption for drying purposes both for processing or for storage (Philippsen et al. 2014), and some authors have proposed more efficient alternatives, such as vacuum drying (Cheng and Torii 2013) or integration of on version through gasification, and power generation in a combined cycle (Aziz 2016).

One of the most effective methods for producing fuel from biomass is pyrolysis, which is performed under oxygen-limited conditions and at a lower temperature, resulting in higher net calorific values (10–20 MJ m^{-3}) than gasification and combustion (4–15 MJ m^{-3}). Pyrolysis yield three final products: bio-oil (30–75%), solid residues or biochar (10–35%), and non-condensable gases (10–35%), such as carbon monoxide, carbon dioxide, hydrogen, and light hydrocarbons (Kositkanawuth et al. 2017). Pyrolysis of seaweeds offers comparable oil yields to pyrolysis of terrestrial biomass. However, compared to conventional liquid fuels, bio-oil has lower quality, derived from the low heating value, high oxygen content, high acidity and viscosity, thus requiring upgrading and refining to improve the quality. Several studies have conducted co-pyrolysis between different materials. Thermal decomposition occurs in a series of stages, decomposition of lipids and carbohydrates, followed by the main decomposition of carbohydrates and then proteins (Han et al. 2018). The high amounts of metals can cause operating problems during the combustion process although they can also act as catalysts (Bae et al. 2011; Han et al. 2018), and the type of catalyst has important effects on the properties of the pyrolysis products (Lee et al. 2014). Several papers have reported that sulfur present in algal biomass has an adverse effect on metal catalysts (Harun et al. 2014).

Thermochemical conversion via pyrolysis was viable to produce energy from a variety of substrates including green tides (Ceylan and Goldfarb 2015) and processing waste products of seaweeds (Poo et al. 2018) and also for co-pyrolysis of *Sargassum* and polystyrene (Kositkanawuth et al. 2017), *Enteromorpha clathrata* with rice husks (Hu et al. 2018) and for cellulose and seaweed polysaccharides (Cao et al. 2018). A comparison of biooils from seaweed pyrolysis can be found in Chen et al. (2015) and Table 6 shows a summary of heat value and yields for biooils and biochars obtained from pyrolysis of different seaweeds.

Biochar can be used as an adsorbent alternative to activated carbon for heavy metal removal. Sargassum chars obtained during pyrolysis (800 °C, 20 min under a nitrogen atmosphere) were modified by impregnation with NH_4Br and used to capture Hg from flue gas (Yang et al. 2018). The mercury removal was enhanced by increasing pyrolysis temperature, O_2 and NO concentration, and by low concentrations of SO_2 and H_2O. Biochars obtained from Sargassum fusiforme at 500 °C and from Saccharina japonica at 400 °C, showed removal efficiencies higher than 86% and 98%, respectively, higher than those attained with pine wood sawdust at 700 °C. Compared to terrestrial lignocellulosic biomass lesser volatilization occurred, and the relatively higher pH and oxygen-containing functional groups of the solid are a beneficial for

Table 5 Some examples of biohydrogen production yields attained from seaweeds

Feedstocks	Fermentative conditions[a] (pH; T; inoculum type[b] and preparation[c])	H_2 yield (L kg^{-1} TS)	References
Codium fragile	5.5; 35 °C; ADS, H	49	Jung et al. (2011a, b)
Ecklonia stolonifera	5.5; 35 °C; ADS; H	43	Jung et al. (2011a, b)
Gelidium amansii	> 5.3–5.5; 35 °C; ADS; H, A	34–53	Park and Pezzuto (2013), Park et al. (2011), Jung et al. (2011a, b)
Gracilaria verrucosa	5.5; 35 °C; ADS; H	46	Jung et al. (2011a, b)
Hizikia fusiforme	5.5; 35 °C; ADS; H	10	Jung et al. (2011a, b)
Laminaria digitata	5.0–5.5 and 7.4–7.6; 37 °C; ADS; H	26	Guneratnam et al. (2017)
Laminaria japonica	5.5–8; 35–37 °C; ADS; Mc, H or EFT	28–110	Liu and Wang (2014), Jeong et al. (2012), Shi et al. (2011), Park et al. (2009), Jung et al. (2011a, b), Shi et al. (2013)
Padina tetrastromatica	–; 35 °C; ADS; H; AP	16–1000	Radha and Murugesan (2017), Park et al. (2009), Jung et al. (2011a, b)
Porphyra tenera	–; 35 °C; ADS; H; AP	9–16	Park et al. (2009), Jung et al. (2011a, b)
Sargassum sp.	7.0–7.2; –; 37; ADS; H	91	Costa et al. (2015)
Ulva lactuca	–; 35 °C; ADS	10	Park et al. (2009)
Undaria pinnatifida	5.5; 35 °C; ADS; H	13–23	Park et al. (2009), Jung et al. (2011a, b)

[a] Batch experiments otherwise stated; *ASBR* anaerobic sequencing batch reactor

[b] *PC* pure culture, *ADS* anaerobic digester sludge from a waste water treatment plant, *ICS* isolated colonies from sewage sludge

[c] *AP* acid pretreated, *EFT* electric field treatment, *H* heat treatment, *Mc* mechanical treatment

[d] L g^{-1}

adsorption applications (Poo et al. 2018). The increased pyrolysis temperature led to a decrease of the yield of biochar, an increase in aromatics and decreased polarity (Jung et al. 2016).

Examples of using seaweed wastes include pyrolysis of *U. pinnatifida* roots, which are the main waste in farming sites and account for 40–60% of annual production, to produce an eco-friendly alternative fertilizer (Jung et al. 2016). *Kappaphycus alvarezii* solid waste obtained after recovery of sap from freshly harvested alga was proposed for energy application using slow pyrolysis at 500 °C, in a packed bed lime scrubber (Das et al. 2017). *Ulva*, cultivated in wastewater from land-based aquaculture and from blooms were proposed for the conversion into biochar to recycle C and nutrients from eutrophic water into agricultural production. Despite suggested water rinsing to reduce salt and pelletization to increase density, the most effective biochar for C sequestration and soil amelioration was produced from un-rinsed and un-pelletised *Ulva* at 300 °C (Roberts and de Nys 2016). Seaweeds treated with Fe can be processed through slow pyrolysis into Fe-biochar, which has a high affinity for oxyanions. Resulting from this Fe-biochar from *Gracilaria* waste was successfully used for the treatment of complex effluents containing Se, As and Mo, difficult to remove through conventional techniques (Johansson et al. 2016).

Alternatively, the use of microwave heated equipment was proposed for pyrolysis of seaweeds industrial solid waste and yielded chars with good properties as a solid bio-fuel and as a precursor of activated carbon. The bio-gas fraction from microwave pyrolysis presents higher H_2 and CO content, and lower CO_2 and CH_4 than that obtained by conventional pyrolysis (Ferrera-Lorenzo et al. 2014). *Porphyra* biomass pyrolyzed in a laboratory-scale multimode-microwave cavity at 400–700 °C produced 87 wt% gaseous fraction, a value higher than from microalgae and comparable with that of conventional gasification processes (Hong et al. 2017).

Hydrothermal carbonization is a less energy intensive thermo-chemical process performed in a pressure reactor under slightly acidic conditions at temperatures of around 200 °C. This technology produces a hydrochar, suited for biomass with high water content and demineralization caused during the process removed alkali salts and chlorine, thus improving combustion properties. *Sargassum horneri* was processed at 180–210 °C with citric acid producing hydrochar with carbon contents of 37–50% and heating values of 19–25 MJ kg^{-1} (Xu et al. 2013) for batch processing of *L. digitate*, *E. hyperborea* and *A. esculenta* by hydrothermal carbonization at 200–250 °C produced hydrochars with energetic value comparable to that of a low ranking coal. The water phase, with high levels of soluble organic carbon, sugars and organic acids, and high levels of K, Mg and P showed potential for production of biomethane and recovery of nutrients following anaerobic treatment (Smith and Ross 2016).

Hydrothermal liquefaction of wild seaweed at 150–200 °C for 1 h, dissolved 55% of seaweed and produced hydrochars, with concentration of potentially toxic elements under quality compost thresholds, except for arsenic (Løes et al. 2018). More dry matter was dissolved by increased temperature, but the solubility of P in chars decreased with higher temperature. The use of organic solvents, methanol and ethanol, at 280 °C increased the yield of bio-oil from *Sargassum tenerrimum* compared to the hydrothermal liquefaction using water at 260–300 °C (Biswas et al. 2017). In comparison to fast pyrolysis hydrothermal liquefaction produced higher yield of bio-oil with more aromatic substances and lower yield of bio-char for *Ulva clathrata* (Hu et al. 2018). Coprocessing wood slurries in hydrothermal liquefaction with brown seaweeds prevented wood slurries dewatering, improving stability and pumpability (Sintamarean et al. 2017).

Gasification consists on subjecting biomass to a high temperature range (700–1000 °C) with either air, hydrogen, oxygen, carbon dioxide, steam, or a mixtures of these to produce a combustible gas mixture called syngas (synthetic gas). Syngas is mostly composed of carbon monoxide, hydrogen, carbon dioxide, minor amounts of methane and trace gases. Steam gasification of algal biomass requires the presence of a catalyst for reforming of the tar derived from algae pyrolysis (Duman et al. 2014). Application of biomass ash as a catalyst improved the gasification rate aiding in the effective utilization of waste ash as well as for the reduction of cost. This strategy was proposed to enhance the gas production from brown

Table 6 Pyrolysis yield and heat value of biooil (BO) and (biochar (BC) from seaweeds

Feedstocks	Yield (%) at 500 °C	HV (MJ kg^{-1})	References
Ascophyllum nodosum	BC: 21.4	21.2	Yuan and Macquarrie (2015a, b)
Cladophora glomerata	BO: 30; BC: 40		Norouzi et al. (2016)
Enteromorpha clathrata	BO: 41.2–45.0	12.0–12.1	Wang et al. (2013, 2018)
Cystoseira barbata	BO: 0.32; BC: 0.21		Cioroiu et al. (2018)
Fucus serratus	BO: 11.0	32.5	Yanik et al. (2013)
Gracilaria gracilis	BO: 65.0; BC:28.0	BC: 13.6	Francavilla et al. (2015)
Laminaria digitata	BO: 17.0	23.1	Yanik et al. (2013)
Laminaria japonica	BO: 29.4–37.5	10.3–33.5	Bae et al. (2011), Lee et al. (2014), Han et al. (2018), Choi et al. (2015)
Porphyra tenera	BO: 47.4	29.7	Peterson et al. (2008)
Saccharina japonica	BC: 31.5		Poo et al. (2018)
Sargassum fusiforme	BC: 38.3		Poo et al. (2018)
Sargassum natans	BO: 33.7	8.7	Wang et al. (2013)
Seaweed mixture	BC: 50.0	15.3	Tag et al. (2016)
Ulva ohnoi	BC: 50.0		Roberts and de Nys (2016)
Undaria pinnatifida	BO: 39.5 BC_{400}: 67.7	23.3	Bae et al. (2011), Jung et al. (2016)

seaweeds (Rizkiana et al. 2014). Also for *Cladophora glomerata*, due to the inorganic compounds in the hydrochar and its porosity (Safari et al. 2016).

The different composition of *seaweeds* has strong implications in the biorefinery process and this is also true for gasification. Seaweeds can generate a net energy of 11,000 MJ/t d. wt whereas microalgae can provide 9500 MJ t^{-1} (Chen et al. 2015; Adam et al. 2015). An integrated power-generation system using *Fucus* sp. as a fuel source, consisting of drying, gasification, and power generation in a combined cycle showed a high efficiency of 60% (Aziz 2016). Brown seaweed, with higher ash content than land biomass, yielded higher gas production (especially for H_2 and CO_2) when gasified with steam in a fixed-bed reactor under atmospheric pressure (Kaewpanha et al. 2014).

Catalytical supercritical water gasification of *L. hyperborea* produced hydrogen and methane, the gas yields increased with temperature, catalyst loading and reaction time The catalyst type strongly influences the product distribution, whereas Ru/Al_2O_3 produced highest yields of methane, sodium hydroxide produced highest yields of hydrogen (Cherad et al. 2014). Seaweeds gasified in supercritical water (500 °C, 1 h) offered coke yields lower than those produced with lignocellulosic and protein wastes with hydrogen yields of 12–16 g H_2 kg^{-1} seaweed and methane yields in the range 39–104 g CH_4 kg^{-1} seaweed (Schumacher et al. 2011).

Gasification has also been proposed as a previous stage before bioconversion with anaerobic microorganisms, either autotrophic or unicarbonotrophic, that transform syngas to bioethanol, but also to other different end products such as hydrogen, butanol, acetic acid, butyric acid, methane or biopolymers. A requisite is that the raw syngas from gasification is cleaned in order to remove compounds that can affect fermentation (Harun et al. 2014; Xu et al. 2011). The potential bacteria (biocatalyst) for syngas fermentation include those of the genus Clostridium, and also some *Acetobacterium* sp., *Butyribacterium* sp., *Methanosarcina* sp. and *Rhodospirillum* sp. were reported (Harun et al. 2014). Biofuels can also be produced by syngas fermentation via a microbial catalysts or via metal catalysts. Syngas fermentation can overcome some disadvantages of the catalytic process, including the need for expensive metal catalysts, susceptible of poisoning and the requirement for severe operation conditions.

3.7 Adsorption

Increasing interest on low cost biosorbents is based on their availability, wide distribution, and renewability. These materials are economically advantageous over industrial synthetic adsorbents, and have great potential for the removal of toxic metals from industrial effluents (Fiset et al. 2008). Intertidal seaweeds are low cost and available adsorbents with potential to eliminate pollutants, such as heavy metals and colorants from waste water. Brown seaweeds are good sorption agents for heavy metal ions, it has been suggested that alginate has a high affinity for biosorption of metal ions and the biosorption capacity is directly related to the presence of binding sites on this polymer (Fiset et al. 2008; Romera et al. 2007). Different behavior among seaweeds have been found, whereas adsorption capacities of 140 mg Pb g^{-1} were found for brown 50–70 mg g^{-1} green seaweeds and 10–40 mg g^{-1} red seaweeds, these latter were the best sorbents for As (Beolchini et al. 2009). Some information is summarized in Table 7.

The application as sorbent can be particularly attractive for invasive species. *Caulerpa racemosa* dried biomass showed a maximum adsorption capacity for methylene blue of 5.23 mg g^{-1} (Cengiz and Cavas 2008), *Sargassum muticum* was efficient for phenols and a chemical pre-treatment with $CaCl_2$ improved the stability and sorption capacity (Rubín et al. 2006). *Sargassum* sp. biomass was suitable for the recovery of ionic copper, the efficiency being different from copper nitrate, copper chloride or copper sulfate solutions (Padilha et al. 2005; De França et al. 2006). Similar behavior for Cd and Cu uptake was observed for three different species of Sargassum biomass (Volesky et al. 1999). Biosorption of dyes was also described, i.e. methyl blue, fuchsin acid, rhodamine B, methylene blue, bromocresol purple and methyl orange, safranin, malachite green and Janus green B onto *Sargassum ilicifolium* (Tabaraki and Khodabakhshi 2017), and methylene blue onto *Gelidiella acerosa* biochar (Padilha et al. 2005; Ahmed et al. 2019). Seaweed collected from beach cast was the cheapest sorbent for phthalates from aqueous solutions and biomass from *S. siliquastrum* showed the highest adsorption capacity. However, the level of desorption efficiency with methanol for beach cast seaweed was higher, although the regenerated adsorbent possessed lower adsorption efficiency than the original biomass (Julinová and Slavík 2012).

Seaweed wastes could be even a cheaper source for this purpose. De-alginated seaweed (the waste from the commercial alginate production) was used for different metal ions, particularly for cadmium (Williams and Edyvean 1997; Romero-Gonzalez et al. 2001), and also Zn and Cr (Cardoso et al. 2017). Consequently the waste of the extraction process, carried out in two stages was successful as bioadsorbent with the same constituents and the same potential to remove toxic metals (Costa et al. 2016). Different brown seaweeds, *Ecklonia maxima*, *Lessonia flavicans* and *Durvillea potatorum*, alginate fibers and de-alginated seaweed waste successfully removed a range of heavy metals (Cu, Ni, Pb, Zn and Cd) (Aderhold et al. 1996). The residue from alginate extraction can be used as adsorbent for metals, the residue from *S. muticum* for Cr(VI) (Belattmania et al. 2017) and from *Sargassum filipendula* for the removal of nickel ion (Moino et al. 2017).

The waste seaweed slurries after the ethanol fermentation with Saccharomyces cerevisiae of the thermal acid hydrolyzed and enzymatically saccharified *Gelidium amansii*, *Gracilaria verrucosa*, *K. alvarezii* and *Eucheuma denticulatum* were reused for the adsorption of Cd(II), Pb(II) and Cu(II) (Sunwoo et al. 2016).

Modification and preparation of seaweeds into stable packing materials to remove heavy metals has been proposed. Continuous metal ions biosorption from Pb and Cu solutions onto *Gelidium sesquipedale* and onto the composite material prepared from an industrial algal waste was performed in a packed bed column using 0.1 M HNO_3 as eluent (Vilar et al. 2008). Different studies confirmed the potential of seaweed wastes operating in a continuous mode in a fixed bed, i.e. the protonated S. muticum biomass for Cd adsorption (Lodeiro et al. 2006) and *Sargassum* sp. for the recovery of ionic copper in a highly stable continuous four column system (Padilha et al. 2005; De França et al. 2006). The operation in several cycles was also feasible for *Sargassum filipendula* alginates wastes used for the removal of nickel (Moino et al. 2017), and the raw seaweed for the removal of Cu (Volesky et al. 2003). Chemically modified de-alginated *Ascophyllum nodosum* was used for the selective removal of the metals (Wolfram, Molibdenum and Vanadium) oxoanion and the composite sorbents exhibited high stability during more than 25 sorption cycles (Mištová et al. 2010). Immobilized particles formulated with *Sargassum thunbergii*, sodium alginate solution, polyvinyl alcohol and activated carbon were proposed for heavy metal adsorption from wastewater in a laboratory-scale fluidized bioreactor operating continuously with mine drainage waste, in repeated cycles, without affecting the adsorption capacity (Li et al. 2017). Other retention procedures include coating *Sargassum muticum* with hydroxides succeeded in removing as from water (Vieira et al. 2017). Combination with clay was tried with *Sargassum* sp. for improving hexavalent chromium biosorption, due to sites to capture and bind the metal ions created by cross-linkage of organic functional hydroxyl groups present in brown seaweed (Aprianti et al. 2017). Another alternative is the use of seaweeds for the preparation of activated charcoal, the direct conversion of *Sargassum fusiforme* biomass to activated carbons in one step was proposed as a simple, lower cost and more sustainable synthesis route (Balahmar et al. 2017). Prior to activation with KOH as an activating agent, biomass sources have to be enriched to carbonaceous matter via hydrothermal carbonization or pyrolysis and the final product showed textural properties and surface functionality similar or superior to analogous carbons prepared via conventional methods. *Ulva prolifera*-based biochar was used to produce a N-doped carbon adsorbent prepared through a rapid hydrothermal carbonization and was used for the adsorptive removal of bisphenol A. The adsorption capacity reached 84 mg g^{-1} (Lu et al. 2017).

A more sophisticated process consisted on the sustainable and cost-effective approach for the preparation of functionalized graphene nanosheets directly from the solids remaining after the extraction of sap from fresh *Sargassum tenerrimum*. Eutectic solvents were employed as solvent and catalyst for the large scale preparation of metal oxide functionalized graphene nanosheets, for the removal of fluoride from groundwater to be used for drinking purposes (Sharma et al. 2017).

Table 7 Adsorption capacity of seaweeds for metals

Seaweed	Maximum adsorption capacity (mg g^{-1})	References
Ascophyllum nodosum	Cd(II) 87.7; Cu(II) 58.8; Ni(II) 43.3; Pb(II) 178.6; Zn(II) 42	Romera et al. (2007)
Asparagopsis armata	Cd(II) 32.3; Cu(II) 40.5; Ni(II) 17.1; Pb(II) 63.7; Zn(II) 21.6	Romera et al. (2007)
Caulerpa racemosa	Methylene blue 5.23	Cengiz and Cavas (2008)
Cladophora fascicularis	Cu(II) 102.3; Pb(II) 198.5	Deng et al. (2006)
Chondrus crispus	Cd(II) 75.2; Ni(II) 37.2; Pb(II) 204; Zn(II) 45.7	Romera et al. (2007)
Codium vermilara	Cd(II) 21.8; Cu(II) 16.9; Ni(II) 13.2; Pb(II) 63.3; Zn(II) 23.8	Romera et al. (2007)
Cladophora fracta	Pb(II) 61.4	Zeraatkar et al. (2016)
Cladophora sp.	Pb(II) 45.4	Lee and Chang (2011)
Cladophora crispate	Zn(II) 31.06	Özer et al. (2000)
Fucus spiralis	Cd(II) 114.9; Cu(II) 70.9; Ni(II) 50; Pb(II) 132–204; Zn(II) 53.2	Romera et al. (2007), Filote et al. (2019)
F. vesiculosus	Cd(II) 125.9; Ni(II) 46.95; Pb(II) 211–259	Rincón et al. (2005), Mata et al. (2016)
Gracilaria sp. biochar	Mo (VI) 78.5; As (V) 62.5; Se (VI) 14.9	Johansson et al. (2016)
Hizikia fusiformis biochar	Ni (II) 12.1, Zn (II) 22.2; Cu(II) 2.2; Pb(II) 2.9; Cd (II) 22.0	Shin (2017)
Laminaria japonica	Al(III) 75.3; Cd(II) 136; Cr(II) 94.1; Cu(II) 101.0; Pb(II) 251–348; Zn(II) 56.9	Lee et al. (2004), Luo et al. (2006)
Saccharina japonica biochar	Cu(II) 93.2–98.6; Cd(II) 60.7; Zn(II) 84.3	Poo et al. (2018)
Sargassum fusiforme biochar	Cu(II) 93.2–98.6; Cd(II) 60.7; Zn(II) 84.3	Poo et al. (2018)
Sargassum ilicifolium	Synthetic colorants 240 mg g^{-1}	Tabaraki and Khodabakhshi (2017)
Sargassum muticum	As(III) 4.2; As(V) 7.3; 2-Clphenol 79; 4-Clphenol 251; Cr(VI) 34.8	Rubín et al. (2006), Belattmania et al. (2017), Vieira et al. (2017)
Sargassum filipendula	Cu(II) 38	Volesky et al. (2003)
Sargassum sp.	Cd 74–87; Cu(II) 51–110; Zn (II) 184	Volesky et al. (1999), Padilha et al. (2005), De França et al. (2006), Karthikeyan et al. (2007)
Sargassum siliquastrum	Phtalates 6.54	Julinová and Slavík (2012)
Ulva lactuca	Cd(II) 29.2; Cu(II) 73.5; Hg(II) 149.25; Pb(II) 34.7	Karthikeyan et al. (2007), Sarı and Tuzen (2008), Zeroual et al. (2003)
Undaria pinnatifida biochar	Phosphate 32.6	Jung et al. (2016)

4 Examples of biorefinery cascading processes

The biochemical characterization of seaweeds is important to identify the best valorization routes. There are huge differences among red, brown or green seaweeds in their composition and depending on the phyla, family, genus or even species level, environmental aspects, seasonality and the processing technologies, the extraction of different fatty acids, oils, natural pigments, antioxidants, and other compounds can fastly differ. Seasonal variations have to be considered when the utilization of invasive species is proposed and also for utilization of industrial by-products, such as the remaining pulp after extraction of high value polysaccharides, i.e. the extraction of alginates from *Sargassum wightii* showed maximum alginate content in March and the leftover pulp was maximum in July, and presented almost 50% insoluble carbohydrates and 30% ash (Kumar and Sahoo 2017).

Seaweeds are commercially used for obtaining high value added products, including hydrocolloids, food, cosmetics, therapeutics and fertilizers. During processing of seaweed leftovers and waste products are generated. They are usually disposed off, however the seaweed biorefinery concept presents a model for the efficient use of the raw material, both for high value added products and the low quality left over biomass for the production of biofuels and energy. A cascading approach promotes that products with the highest value is made first, and those with lower value are made after, thus bioenergy and biofuels are preferably obtained from leftovers from other production processes. This approach increases resource efficiency (Balina et al. 2017). Biorefinery is based on the circularity principle, considering reduced or no waste, product reusability and recyclability, and providing energy from renewable and sustainable sources. Seaweed biorefinery can also be addressed to use seaweed species that cause blooms as a result of eutrophication preventing negative ecological impact and improving the sustainability of coastal environments. This approach minimizes the amount of waste produced to a nearly zero waste system, thus strengthening the global competitiveness of biobased industries and giving the highest social, environmental and economic benefits (Goh and Lee 2010; Balina et al. 2017).

The focus lies on the integral utilization of valuable components, despite that each raw material can be more suited for a particular end-use. For example, a study to determine the capacity of seaweeds to produce value-added-molecules for food, feed, cosmetics, nutraceuticals and energy taking into account their biochemical characteristics it was found that *Palmaria palmata* was best suited for food and for anaerobic digestion (0.279 LCH_4 g^{-1} VS) while *Saccharina latissima* could be used for alginate extraction (242 g kg^{-1}, M/G = 1.4) and *Sargassum muticum* for polyphenol extraction (20 g kg^{-1}) (Jard et al. 2013).

4.1 Brown seaweeds

From a biorefinery point of view, brown seaweeds are very interesting (Veide Vilg and Undeland 2017). During the manufacture of alginic acid from seaweeds, filter cake and the final filtrate are discarded as waste, which still contain 93–94% of the iodine present originally in the seaweeds, and could be regarded as a potential source of iodine (Dave and Sharma 1974) More recently, sequential utilization of the saccharidic fractions and other components has been proposed.

The acidic treatment was applied for the sequential extraction of fucoidans and alginates from brown seaweeds such as, *E. radiata*, *M. pyrifera*, *D. potatorum*. The cascading process allowed the extraction of 30–40% of total available fucoidans and 80–94% of total available alginates. The fucoidan extracts had up to 30% sulfate and up to 30% fucose and the presence of phlorotannins contributed to the antioxidant properties (Lorbeer et al. 2017).

These authors proposed a combined alkaline extraction and acid precipitation in a simple, scalable, pH shift-based process applicable on wet biomass to recover *S. latissima* proteins. Protein could be used as a highly desired vegan protein for human consumption or applied in animal feed as protein. Ethanol production from seaweed with integrated production of protein-rich ingredient for fish feed and liquid fertilizer from offshore brown seaweed cultivation contribute to environmental restoration and climate mitigation. Bioextraction of nitrogen using seaweed cultivation as biofilter reduces marine eutrophication by 16.3 kg N eq ha^{-1}. The cost of biomass productivity through seaweed cultivation is the main constraint for competitive prizes with other energy and protein producing technologies (Seghetta et al. 2016a, b, c).

An invasive seaweed species is *Sargassum muticum*, see (Kraan 2008) for a review of arrival of this species at several European shores. *Sargassum muticum* was processed with conventional technology to extract alginates, membrane technology was accomplished to recover phenolic fractions and the solids were fractionated with hydrothermal technology to solubilize the mineral, fucoidan and phenolic fractions; the remaining solids showed lower calorific power than wood, but could be proposed as soil amendment for their mineral content (Fig. 1a) (Pérez et al. 2014; González-López et al. 2012) proposed the microwave processing of *S. muticum* to obtain an aqueous extract with antioxidant properties and the solid phase remaining could be further processed for the extraction of fucoxanthin with higher yield than from the freeze dried seaweed (Balboa et al. 2015; Conde et al. 2015). Alternatively, the solids could be further processed by enzyme assisted extraction or hydrothermal treatment (autohydrolysis), this treatment allowed the simultaneous extraction and depolymerization of fucoidans (Balboa et al. 2013a, b), or the successive depolymerization (Flórez-Fernández et al. 2017a, b, c) of compounds with antiradical and antitumoral properties. The phenolic fraction found in the liquid phase could be further recovered and concentrated with nonionic polymeric resins and the dried product showed antioxidant and anti-inflammatory properties (Casas et al. 2016). In a different scenario, fucoidans and phenolics from the liquid phase obtained in the autohydrolysis step could be selectively concentrated using ultrafiltration membranes operating in diafiltration mode, lowering the mineral content (Balboa et al. 2015).

Cooking liquid, remaining after *Hizikia fusiformis* processing in the food industries, contain large amount of protein, carbohydrates, and phenolic compounds. After irradiation, which can be used to inactivate pathogens, the color of the cooking liquid became brighter. It showed that the amount of ethanol extractable total phenolics increased, regardless the type of irradiation source, i.e., gamma rays or electron beam. Consequently, the antiradical properties were enhanced and there was significant increase in the tyrosinase-inhibiting and angiotensin L-converting enzyme inhibitory activity which could have applications in cosmetics (Kim et al. 2009; Choi et al. 2011).

Quitain (2013) processed the supercritical carbon dioxide deoiled *Undaria pintatifida* by hydrothermal treatment to recover and degrade fucoidan into highly potent low-molecular-weight components (5–30 kDa). Microwave heating showed advantages over conventional heating, obtaining the target molecular weight at a temperature close to 140 °C and short irradiation time (1 min).

Extraction of alginate from food and cosmetics applications is a commercial use of brown seaweeds, but an additional utilization of sodium alginate at 1% (w/v) and calcium nitrite at 11.3% (v/v) could be corrosion inhibition. Cactus mucilage and brown seaweed extracts are low-cost corrosion inhibitors for steel in concrete, reducing the use of biohazards and toxic inhibitors (Hernández et al. 2017). Sodium alginate has also been used as a biodegradable additive in the papermaking industry. The addition of sodium alginate alone had little effects on the paper properties, but addition of 0.2% sodium alginate prepared from waste seaweeds and coupled with polyamideamine–epichlorohydrin could increase the contact of fibers in paper sheets causing an enhancement of the mechanical properties (dry tensile index, wet tensile index, tearing index, bursting index, and folding endurance) of paper from recycled fibers, compared with addition of 0.8% PAE alone (Bai et al. 2017). A bio-refinery approach using Laminaria digitata to obtain fucoidan, alginate and ethanol (Fig. 1b) was proposed (Kostas et al. 2017). The process consisted on an initial extraction with 0.1 M HCl at 70 °C, after filtration to separate the extract, alginate was precipitated with $CaCl_2$ and in the alginate-free liquor, and ethanol was used for precipitating fucoidan, isolated with a purity of 65%. The remaining residue, mainly consisting on crude fiber, with glucose as the predominant monosaccharide, was hydrolyzed with dilute sulfuric acid hydrothermal pre-treatment (1.5 N H_2SO_4, 24 min, 121 °C) and enzymatic saccharification yielded 93.8% of theoretical glucose.

In a study for kelp biorefinery into biogas and fertilizer, also harvesting and anaerobic digestion were identified as the stages requiring optimization to have a large impact on scale up processes and on energy use and emissions. Emissions and energy demand could be almost halved by scaling operations up by a factor of twenty (Pechsiri et al. 2016).

4.2 Green seaweeds

Different studies confirmed the opportunities to deliver multiple products through a biorefinery process from green seaweeds, such as from *Derbesia tenuissima* and *Ulva ohnoi* (Mata et al. 2016). A valorization strategy was proposed of Ulva biomass as feedstock for several products using a cascading biorefinery approach consisted on the solubilization of sugars by hot water followed by enzymatic hydrolysis to obtain a sugar-rich hydrolysate containing glucose, rhamnose and xylose, and a protein enriched fraction (34% wt) (Fig. 2).

The extracted protein fraction showed improved value as animal feed ingredient due to the increased amino acid content. Ileal digestibility and rumen fermentation where much better compared to whole *Ulva*. The hydrolysate was used successfully for the production of acetone, butanol, ethanol and 1,2-propanediol by clostridial fermentation (Bikker et al. 2016). Effective recovery of Ulva lactuca constituents, including crude proteins with digestibility of protein of 86%, was reported from fresh biomass after extraction of sap (Gajaria et al. 2017).

An integrated biorefinery was proposed for the co-production of bioethanol and biogas from *Chaetomorpha linum* (Ben Yahmed et al. 2016). A pretreatment with 3% NaOH (120 °C, 1.5 bar) gave the best result in terms of thallus disintegration, biomass recovery

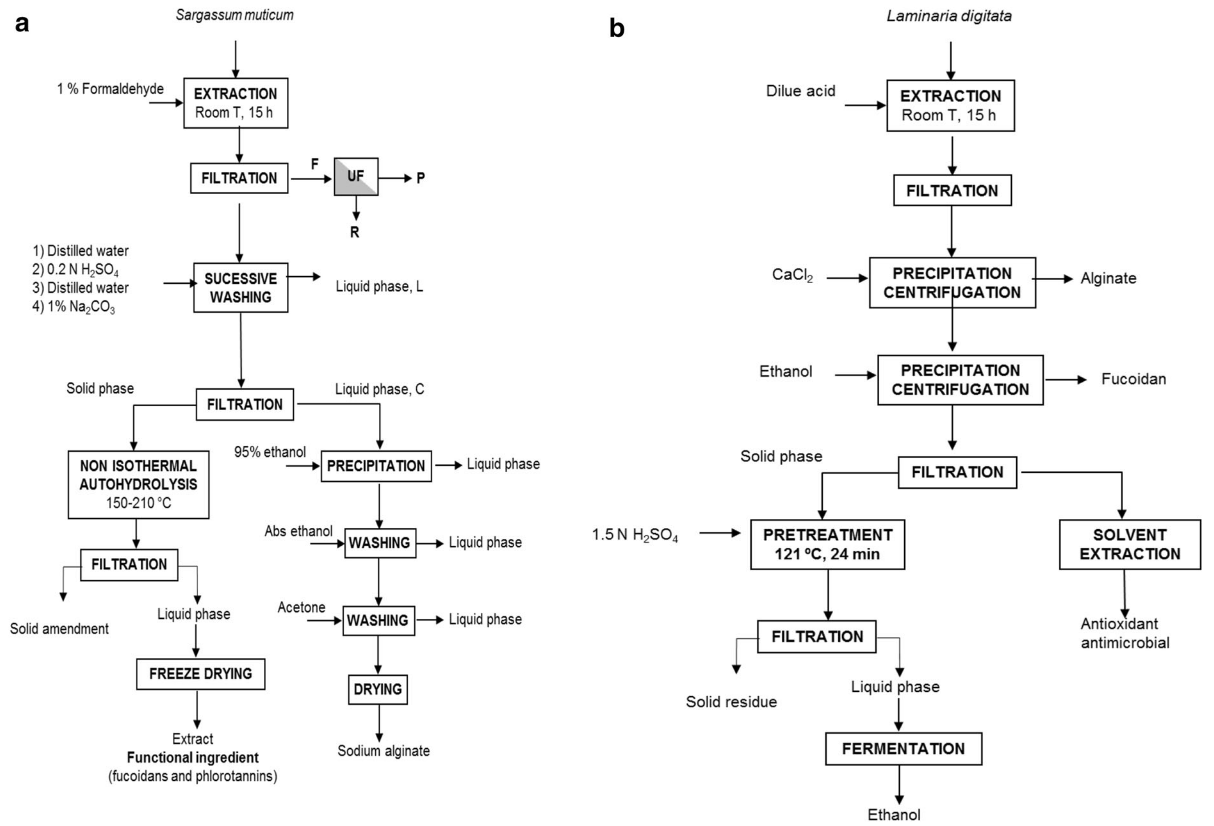


Fig. 1 Cascade processes for the integral valorization of **a** *Sargassum muticum* (González-López et al. 2012) and **b** *Laminaria digitata* (Kostas et al. 2017)

and digestibility with a cocktail of cellulases, hemicellulases, proteases,…produced by *Aspergillus awamori*, growing on the alga and recovered by ultrafiltration (10 kDa). An ethanol yield of 0.41 g g^{-1} sugar, corresponding to 0.09 g g^{-1} pretreated alga was obtained after fermentation by *S. cerevisiae*. Different waste streams, including the mycelium from the fungal fermentation, the residual non-hydrolysable biomass and all effluents including the gas, CO_2, discharge from the fermentation and the vinasse residue after distillation, were jointly valorized for anaerobic digestion to biogas, with a biomethane yield of 0.26 L g^{-1} volatile solids (VS). The process was not inhibited by minerals and sulfated sugars since they were diluted. Although the seasonal variability and environmental constraints could affect the chemical composition of the seaweed and the yields, the process provided comparative yields of bioethanol and biomethane from other green seaweeds. An economic assessment confirmed that biomass feedstock and its transportation accounted for 14%, hydrolysis enzymes for 22% of the cost, whereas the high energetic efficiency was related to the production of bioethanol and biogas, and to the energy saving related to the absence of seaweeds cultivation and transportation.

A novel approach consisted on the recovery of crystallized seaweed salt after evaporation of the water used for washing of biomass of *Ulva ohnoi* and *Ulva tepida*. Washing produced Na:K salts that could be used for food applications, and maximum yields were 29% for *U. ohnoi* and 36% for *U. tepida*, and up to 19% salts for ulvan production. This stage also reduced algal mineral content, thus improving the quality of the biomass for utilization as fertilizer, feed or fuel, increasing the energy content of the biomass by 20–50% to a maximum of 18 MJ kg^{-1} and protein contents by 11–24% to a maximum of 27.4% (Magnusson et al. 2016). Furthermore, Glasson et al. (2017) proposed a cascade biorefinery for *Ulva ohnoi* to obtain salt, ulvan, and a protein enriched residual biomass for animal feed, chemicals and biofuels. The sequential process consisted of aqueous extraction of salt, ethanol extraction of pigments, and $Na_2C_2O_4$ or 0.05 M HCl extraction of ulvan, this latter process provided higher total sugars and ulvan yields (8.2% wt/wt) and purity than the first (4.0% wt/wt).

4.3 Red seaweeds

The utilization of agarophytic feedstocks for biorefinery has been frequently described. In addition to the components present in other types of seaweed, red seaweed contain natural colorants (R-phycoerythrin, R-phycocyanin) and higher proportion of protein, which could be proposed for biotechnological, nutraceutical and pharmaceutical applications (Francavilla et al. 2013). Therefore, the sequential extraction of the major components of red algal biomass would be slightly different, aimed at obtaining products such as pigments, lipid, agar, minerals and energy dense substrate (cellulose).

A **cascade biorefinery** approach of *Gracilaria gracilis* consisted on the extraction of phycobiliproteins [R-phycoerythrin (7 mg g^{-1} d.w.), allophycocyanin (3.5 mg g^{-1} d.w.) and phycocyanin (2 mg g^{-1} d.w.)] and further pyrolysis of the residue to produce bio-oil and biochar. The bio-oil yield was ~ 65 wt% at ~ 500 °C, although the high nitrogen content prevents its use as a biofuel, unless denitrogenation takes place. Biochar yields ranged between 26.5 (600 °C) and 33 wt% (400 °C) and inorganic elements such as P, K, Ca, Fe and Mg were present in the product (Francavilla et al. 2015).

Production of agar and ethanol from *Gracilaria verrucosa*, in a biorefinery approach after selecting the optimal periods for algal harvest, resulted in extraction of 27–33% agar and the leftover pulp contained 62–68% holocellulose, subjected to enzymatic hydrolysis and further fermentation (Kumar et al. 2013).

Baghel et al. (2015) reported the basis for ocean-based bio-industries based on *Gelidiella acerosa*, *Gracilaria dura* and *Gelidium pusillum*. Process scale up was feasible since the yields and properties of all products were nearly the same as those from lab scale. The fresh algal material was subjected to aqueous extraction (phosphate buffer, 12 h, 4 °C) in two stages, centrifuged and the supernatant containing crude phycobilin pigments was collected and purified by ammonium sulphate precipitation. The supernatant was considered as liquid fertilizer. Lipids were extracted from the residues leftover after chloroform–methanol (1:2 v/v) extraction. The residue remaining after lipid extraction was mixed with distilled water (120 °C, 1.5 h) after centrifugation the supernatant was collected and gelled to obtain the native agar. The residual masses remaining after agar extractions were used for cellulose extraction, bleached (36% $NaClO_2$, 60 °C, 8 h), washed with water until neutral pH, treated with 0.5 M NaOH solution (60 °C, 12 h), washed with water until neutral pH, re-suspended in 5% v/v HCl and boiled. Cellulose was hydrolyzed with commercial enzyme cellulose and the hydrolysate was fermented to ethanol.

Another example of a cascade application is a simple benign process based on aqueous extraction of *Gracilaria corticata* with no solid waste generation and allowed the sequential recovery of products including R-phycoerythrin, R-phycocyanin, crude lipid, agar, soil conditioner, bioethanol and mineral rich liquid with possible fertilizer. The following stages were reported; water extraction (12 h, 4 °C) in several stages, filtration to separate the crude pigment which was processed by ultrafiltration (200 kDa) to separate the concentrated pigment mixture (R-phycoerythrin and R-phycocyanin) and mineral rich water (Fig. 3a).

Recyclability of the mineral rich water obtained after ultrafiltration was proposed for pigment extraction from at least two batches or used as plant nutrient rich extract for foliar applications. The dried residue from pigment extraction was used for lipid extraction with chloroform–methanol (1:2 v/v) at room temperature in several stages. Crude lipid extracted from the residual biomass from pigment extraction was comparable to that conventionally extracted from the primary feedstock. The residual biomass after lipid extraction was used for agar extraction with distilled water (120 °C, 1.5 h), the agar was obtained after gelling (−20 °C, 15 h) and thawing, with recovery yields similar for both biorefinery integrated and for the conventional process from primary biomass. The residual pulp remained after agar extraction was dried and hydrolyzed with 2% cellulose with a yield of 0.27 g sugars g^{-1} residue, and S. cerevisiae produced ethanol from hydrolysate with 92% conversion efficiency. The residue remaining after fermentation presented a C:H:N:S composition favorable for

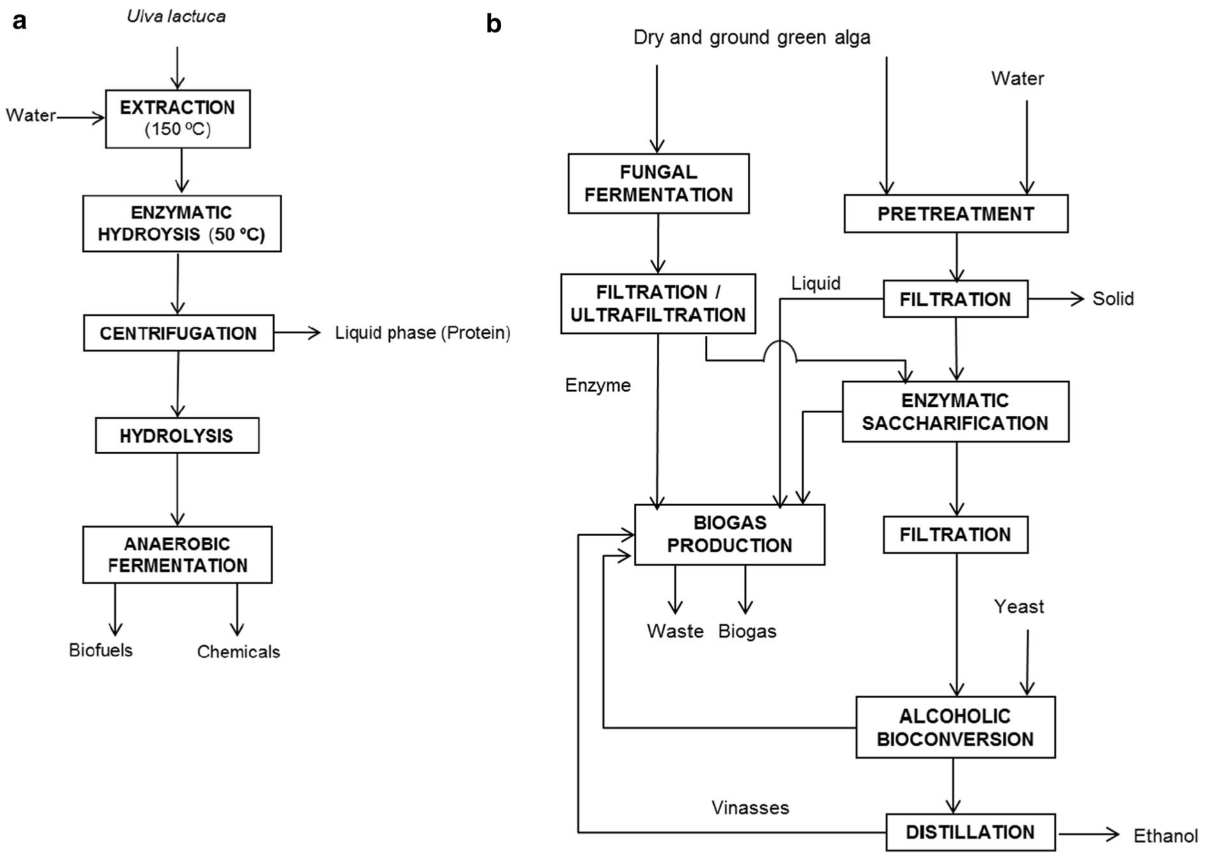


Fig. 2 Cascade processes for the integral valorization of **a** *Ulva lactuca* (Bikker et al. 2016) and **b** other green seaweed (Ben Yahmed et al. 2016)

utilization as soil conditioner. It is estimated from pilot scale trials that one ton fresh biomass yielded up to 0.3 kg crude pigments, 1.8 kg lipids, 28 kg agar, 3.6 kg bioethanol, 15 kg of soil conditioner and up to 610 L of mineral rich liquid fertilizer. If the biomass were processed for recovery of agar alone, the ratio of processing cost and value of product is 1:2 whereas for multiproduct recovery is 1:4 (Baghel et al. 2016).

The industrial utilization for fresh *K. alvarezii* considers the production of a sap rich in potash and micronutrients and a granular residue rich in κ-carrageenan (Mantri et al. 2017). Additional compounds have been considered, including bioethanol, carrageenan, fertilizer, and biogas (Ingle et al. 2018). An integrated scheme for the production different components from *K. alvarezii* was reported. Initially the fresh seaweed was crushed to expel the juice rich in KCl, then κ-carrageenan was extracted from the granular residue and 5-hydroxymethyl furfural was produced using $Mg(HSO_4)_2$ catalyzed reaction, which also produced galactose, which remained in the aqueous phase. The aqueous phase was recycled up to 10 times by maintaining a constant acid strength, and utilized thereafter for the recovery of K_2SO_4. The spent aqueous phase rich in galactose was subjected to further reaction with HCl to yield levulinic acid and formic acid in nearly equal proportions. Combustion/gasification of this biomass would yield additionally 0.74 t fertilizer and the required amount of H_2SO_4 for $Mg(HSO_4)_2$ preparation (Mondal et al. 2013).

Another alternative consisted on the acid hydrolysis/saccharification of κ-carrageenan rich granular biomass, which can be washed with water to remove adhering salt followed by sun drying and used. After saccharification (0.9 N H_2SO_4, 100 °C, 1 h) the yield with respect to κ-carrageenan in granules depend on the operation scale (26–31%, wt/wt). The hydrolysate was neutralized with lime and the filtrate was desalted by electrodialysis and fermented with *S. cerevisiae* for ethanol production with 80% conversion yield; however 50% of the total organic carbon present in the initial biomass remained unutilized (Khambhaty et al. 2012). The economics were more favorable for multi-stream, zero-effluent processing of fresh biomass than for conventionally dried seaweed (Shanmugam and Seth 2018).

Agar and carrageenan from red seaweeds are the hydrocolloids extensively used in the food industry for their gelling, emulsifying, thickening, and stabilizing properties (Pereira et al. 2015). However, the seaweed phycocolloids industries use only 15–30% of the total dry mass, whereas the remaining 70–85% is degraded or drained out as a waste with effluents. The utilization of this waste fraction has been proposed in different studies.

Seaweed by-products generated after phycocolloid extraction can be utilized as a source of protein. Since their extraction requires successive washings with water, the first cold-water washing stream contains many proteins, including phycobiliproteins. This wastewater stream from *Porphyra columbina* processing can be hydrolyzed with proteases in a single stage or in a sequence, and both the protein extract and the hydrolysates showed immunosuppressive, antihypertensive and antioxidant properties (Cian et al. 2012, 2013). Seaweed by-products after agar extraction are good sources of protein and amino acids with low fat content. Gracilaria fisheri protein fraction after agar extraction was hydrolyzed using bromelain (119,325 U g^{-1}, at 10% (wt/wt) for 3 h), with a 38% yield and 63% degree of hydrolysis. The most abundant free amino acids were arginine, lysine, and leucine and the predominant odorant compounds were hexanal, hexanoic acid, nonanoic acid, and dihydroactinidiolide and presented umami taste and seaweed odor. Thermally processed hydrolysates showed roasted seafood-like flavor (Laohakunjit et al. 2014). The residual cake remaining after phycocolloids extraction from Porphyra columbina contains 27% protein and 10.7 mg gallic acid equivalents/100 g. Enzymatic hydrolysis of the residual cake enhanced the protein solubility and the residual cake hydrolysate has low molecular weight peptides containing Asp, Glu, Ala, and Leu with 45% angiotensin-converting-enzyme (ACE) inhibition, chelating capacity and radical scavenging activity against ABTS and DPPH, attributed mainly to low molecular weight peptides (500 Da) and polyphenols compounds (Cian et al. 2013).

Seaweed solid wastes obtained after the extraction of κ-carrageenan from *E. cottonii* were processed by separate enzyme hydrolysis with a cocktail of cellulolytic enzymes and fermentation with Saccharomyces cerevisiae (SHF) process and simultaneous saccharification and fermentation (SSF). In SHF up to 99.8% glucose yield was obtained at pH 4.8, 50 °C and the subsequent fermentation yielded 55.9% bioethanol, whereas the SSF process yielded 90.9% bioethanol. The SSF is advantageous over SHF, since it is a simple one-step procedure saving time, cost and energy consumption achieving a high yield of bioethanol (Tan and Lee 2014).

Anaerobic digestion of waste sludge produced during the industrial extraction of alginate from *Laminaria hyperborea* and *Ascophyllum nodosum* was carried out in bench scale intermittently stirred digesters at 35 °C. Sieve and flotation sludges were digested in batch and semi-continuous cultures. Methane production was recorded in the range 0.10–0.28 L g^{-1} VS (Kerner et al. 1991).

5 Challenges and constraints

Seaweeds are resources that represent a valuable sustainable feedstock either as beach cast, algal blooms or from aquaculture. Over 20 million tons of seaweed is cultivated every year, mainly in the far East (FAO 2018). This biomass can be used for direct human consumption or further processed into food ingredients and additives, pet food, feeds, fertilizers, biofuel, cosmetics, and medicines. The production of valuable components and additives from seaweeds generates several by-products and waste biomass that are usually discarded, however from an economic, social, and environmental perspective this waste biomass is of great interest as it contains valuable bioactives and biomolecules. Seaweed biorefinery systems could offer an opportunity to economically strengthen industrial sectors as agriculture, fishery, chemical and energy, while helping the circular economy, recycling of nutrients and alleviating environmental issues.

The unique chemical composition of seaweeds and their fast growth offer many opportunities for biorefining (Van Hal et al. 2014). Seaweeds contain

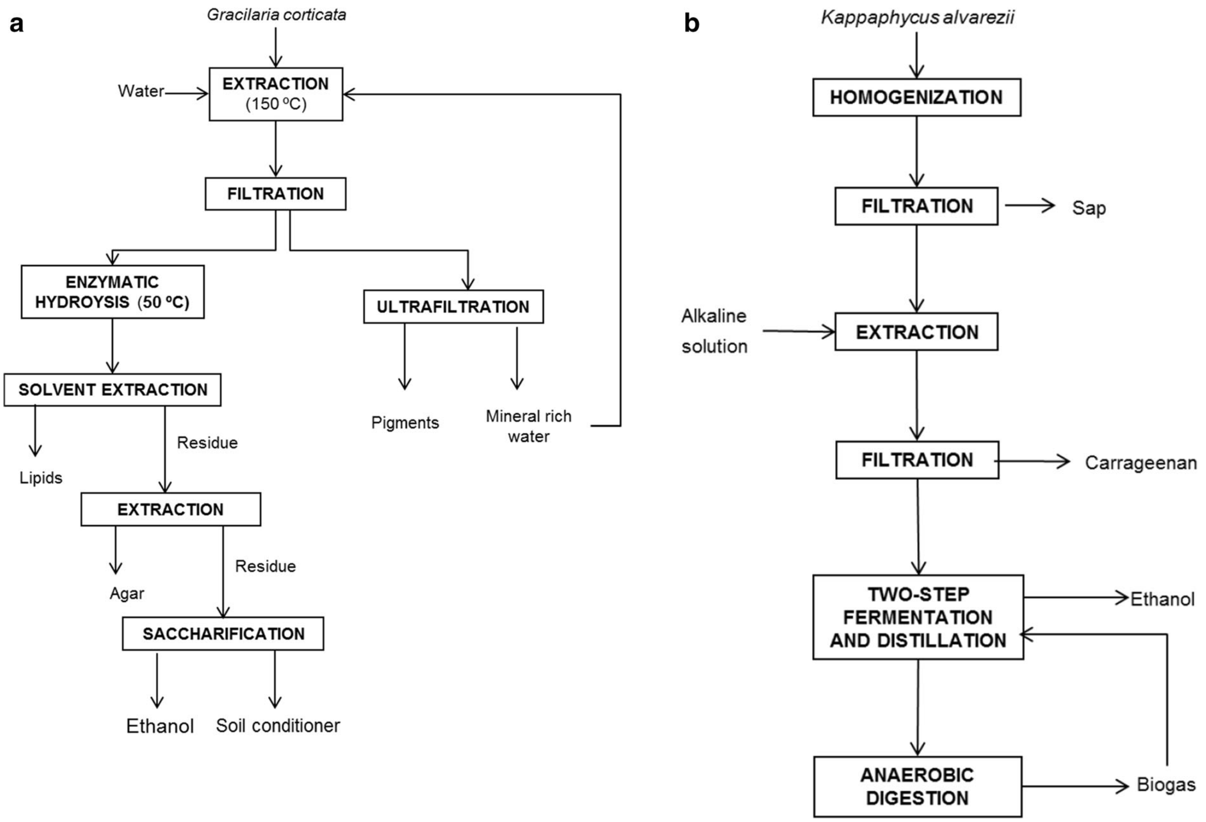


Fig. 3 Cascade processes for the integral valorization of **a** *Gracilaria corticata* and recyclability of water (Baghel et al. 2016) and **b** conversion of red *Kappaphycus alvarezii* (Ingle et al. 2018)

exclusive components different from those of terrestrial biomass, and can be cultivated with currently available farming technology. Cultivation of seaweed can be a profitable activity, especially for coastal communities because it has a short production cycle, low capital requirement, and relatively simple farming technology. In addition, seaweeds can efficiently absorb CO_2 from the seawater. Eutrophication resulting into massive seaweeds growth or algae blooms may be considered as a biomass source for industrial applications and for biogas production. Commercial exploration of invasive species for some uses such as fuel could encourage its harvesting and control. Anaerobic digestion is one of the applications that have been suggested, taking into account seasonal harvesting and, the need to preserve and store seaweed to supply a year-round biomass.

Still many challenges remain with respect to use of seaweeds for chemical production, such as the seaweed availability and large seasonal variation in the chemical and nutritional composition of seaweeds (Van Hal et al. 2014). Some barriers like limited technologies and unpredictable amount and quality of seaweed biomass still exist (Balina et al. 2017). Seaweed biomass can differ among species, geographical areas, season and the yields and type of products obtained are highly dependent on the processing technologies. Generally, the technologies used for processing terrestrial-based biomass are suited and the application of emerging technologies or the development of novel ones could be successful (Jung et al. 2013).

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