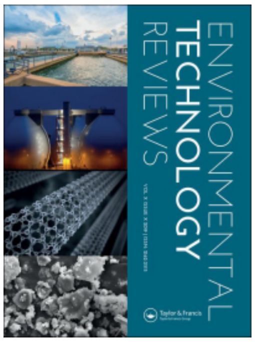

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REVIEW ARTICLE

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Valorisation of macroalgae for biofuels in Indonesia: an integrated biorefinery approach

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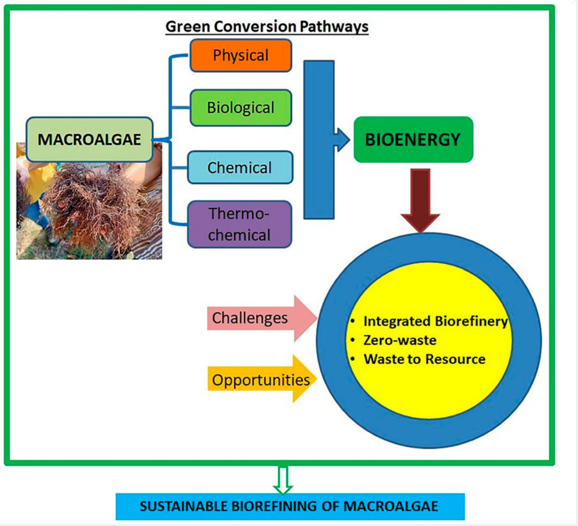
ABSTRACT

Globally, bioenergy development depends on the efficacy and affordability of conversion technologies and the availability of renewable biomass sources. As a tropical country, Indonesia has a wide range of biomass sources, either from land or marine biomass (i.e. macroalgae). The current estimation of macroalgae potential in Indonesia is estimated at approximately 9.96 million tonnes or about 26.86% share of world production in 2021. Specifically, marine macroalgae (wild or cultivated) have received attention for their potential as renewable resources for the sustainable bioenergy production, supporting a move towards a circular economy. However, as a developing country, Indonesia still needs to evolve and further advances its technology and skill capacity to address research, development, and innovation challenges in this area. Thus, this paper examines the potential biorefinery approach for application and commercialisation in Indonesia. It discusses cultivation practices and the future direction of the most sustainable and feasible routes for bioenergy production from macroalgae, exploring recent developments, opportunities, and challenges towards circular processes. The study proposed that the biorefining of macroalgae into bioethanol, biogas, compost, and solid fuels, either as mono – or co-production, are potential. Therefore, this paper may offer to narrowing the literature's gap and adding a new perspective on the adoption of macroalgae-based bioenergy with integrated biorefinery and closed-loop systems approaches.



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Biorefining; circular economy; conversion technology; macroalgae supply chain; macroalgae-based bioenergy

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Highlights

- Macroalgae is renewable and sustainable feedstock
- Seasonal variability and harvesting influence macroalgae quality
- Several scenarios on feasible routes of macroalgae-based biofuels
- Biorefining of bioethanol and biogas from macroalgae creates bioeconomy
- Challenges remain on cultivation technology, regulatory support, and supply chain

List of abbreviations including units and nomenclature

AD	Anaerobic Digestion
ABE	Acetone-Ethanol-Butanol
BMP	Biochemical Methane Potential
BM	Ball Milling
BOE	Barrel of Oil Equivalent
BT	Beating
CFCS	Circulation and Floating Culture System
CSTR	Continuous Stirred Reactor Tank
CV	Calorific Value
FAME	Fatty Acid Methyl Esters
GHG	Greenhouse Gas
GPNE	General Planning for National Energy
HHV	High Heating Values
HRT	Hydraulic Retention Time
HTL	Hydrothermal Liquefaction
HTP	Hydrothermal Pretreatment
IMTA	Integrated Multi-Trophic Aquaculture
LPG	Liquefied Petroleum Gass
MC	Moisture Content
MEMR	Ministry of Energy and Mineral Resources, Republic of Indonesia
MG	Machine Gap
MPBR	Macroalgae Photobioreactor
NEP	National Energy Policy
NRE	New and Renewable Energy
OLR	Organic Loading Rate
SMP	Specific Methane Potential
TS	Total Solids
VS	Volatile Solids
VFA	Volatile Fatty Acid
WW	Wet Weight

1. Introduction

Coal, oil, and gas still account for around 90% of Indonesia's total national primary energy supply [1]. The coal company in Indonesia is enlarging by up to 48.1% from the past to the current condition, which shows the heaviest reliance on this unrenewable energy resource [2]. However, decreasing fossil fuel resources and an increase in electricity demand nationally have led the Indonesian Government to focus more on developing sustainable and renewable energy. Approximately 146.7 million tonnes of biomass are produced in Indonesia annually (470 GJ/y equivalents) with an estimated energy equivalent of 32.656 MW [3], but this does not include marine biomass. The national macroalgae production data from the primary producers reached up to 9.32 million tonnes in 2020 [4] and increased to 9.96 million tonnes in 2021 [5]. This production makes Indonesia as the second-highest macroalgae producer, which contributes approximately 26.86% of the global production (or 35.76 million tonnes in 2021) after China (56.75%) [6]. Macroalgae production is expected to unfold many times since the global population growth is expected to reach up to 70% by 2050 [7]. This data indicates excellent potential and availability of macroalgae for usage as biofuels in Indonesia.

In addition, the final energy consumption for 2020 from biomass is 53.37 million barrel of oil equivalent (BOE) from approximately 32.6 GW of biomass potential covered in Indonesia, which means it still needs more optimum utilisation [8]. Furthermore, energy and food security have been highlighted as two critical issues that need to be addressed by the Indonesian Government to meet the national sustainable development goals (SDGs). Limited access to affordable, sustainable energy and food resources remains one of the most significant challenges faced by the country. Approximately 14.9 million households in Eastern Indonesia rely on traditional biomass sources for cooking (e.g. wood). Access to renewable energy is restricted in rural areas compared to urban areas [9]. Electricity demand is estimated to increase by 1.083 TWh (business as usual model) and 1.193 TWh (electric vehicle model) in 2050 [9,10]. Some of these issues can be alleviated through the sustainable conversion of biomass to energy and food. Therefore, the Indonesian Government has promoted biomass utilisation for bioenergy production as stated in several policies (i.e. No 79/2014 relating to the National Energy Policy (NEP)) with a target of 23% to achieve in 2025 and 2050 [11]. Also, the order of the National Energy General Plan (RUEN) as the Presidential Decree Number 22 of 2017 is stipulated to increase biomass into biofuel account for 22.7 million tonnes in 2050 [10]. Therefore, considering the availability of macroalgae and the support of policies/legislation, valorising this substrate into biofuels is an attractive option.

As a developing country, Indonesia faces numerous challenges in accelerating the broader adoption of sustainable bioenergy and materials from existing biomass resources. One major challenge is access to credible and robust data on the location and nature of these currently disparate resources. Currently, information on efficient routes for conversion and existing supply chains and logistics that may support this is limited [12]. Also, implementing renewable energy technologies is far behind the applicable stage, and the estimated potential is distinct between studies from the research field. Hence, the urge to reassess data and aggregate technologies is needed to tackle those issues [13]. In Indonesia, current technologies and processes' high capital and operational costs, insufficient and inefficient grid infrastructures, land use and availability, and the challenges of utilising lignin-rich biomass are currently barriers to the broader adoption of bioenergy projects [14]. The wider sustainability issues surrounding using purpose-grown crops for non-food bioenergy crops have led many countries to seek alternative biomass sources (e.g. waste). Competition for land and resources and focus on food security have also placed greater attention on marine-based biomass (i.e. macroalgae) [15].

Macroalgae can be categorised into red algae (*Rhodophyceae*), green algae (*Chlorophyceae*), and brown algae (*Phaeophyceae*) [16,17]. These macroalgae can be used as sustainable resources to produce bioenergy (i.e. biogas, biohydrogen, biodiesel, bioethanol, biochars, etc.) [18–20] and bioproducts (i.e. cosmetics, animal feeds, organic acids, medicines, chemicals, etc.) [21–23], with the application of integrated biorefinery concept [24]. Many studies have constantly highlighted the utilisation of macroalgae for biofuel production [5,25]. For instance, biogas production potential from macroalgae ranges from 0.100 to 0.855 m^3 CH_4/kg VS [18,26,27], much higher compared to the cow manure (0.216 m^3 CH_4/kg VS) [28] and maize silage (0.191 m^3 CH_4/kg VS) [29]. Other macroalgae, for instance, *Rhizoclonium sp.* could produce 65.43 ± 18.13 g/L of bioethanol [30], *Padina tetrastromatica* could extract 78 mg/g algal biomass of biodiesel, and *Saccharina japonica* could produce 35 MJ/kg high energy content. A previous study by Loh et al. [31] reported the superior potential of applying the cascading biorefinery approach from *Eucheuma denticulatum* for bioethanol production with the residual macroalgae used for biogas and the produced wastewater for algae cultivation. Furthermore, Dickson and Liu [32] reported that the biorefinery approach of *Saccharina japonica* for bioethanol production while utilising all waste streams generated from the process has a high economic feasibility, a lower risk probability (i.e. 20–44%), and 90% reduction of environmental impacts. A current study by Arias et al. [33] also pointed out the application and commercialisation of multi-products biorefinery of macroalgae to produce biogas and bioproducts (i.e. lipids, protein, carrageenan, and biostimulants). Moreover, Kumar et al. [34] investigated the biorefinery integration of red algae (*Gracilaria verrucosa*) for agar extraction and ~68% of the leftover pulp for bioethanol production. These studies demonstrated strongly that macroalgae for bioenergy promote great opportunities for sustainable and commercially viable stages.

As an archipelago country, Indonesia varies in geospatial distribution and types of macroalgae, climate, and infrastructure (i.e. water, energy, and waste management). In addition, Indonesia still faces problems with capabilities and capacities in research and development, construction and operation of renewable technologies, and access to finance and investment. Therefore, this paper explores and compares macroalgae valorisation for bioenergy, pretreatment, conversion process, scaling-up, and commercialisation globally and specifically in the Indonesian context. It discusses cultivation practices and the future directions of the most sustainable and feasible routes for bioenergy production from macroalgae, exploring recent developments, opportunities, and challenges towards circular processes. The promising routes of the biorefinery approach for application and commercialisation in Indonesia are presented and evaluated. These are based on peer-reviewed studies and consider macroalgae availability, valorisation scenarios, and technological, economical, and environmental factors. There are limited studies or comprehensive reviews on the prospective adoption of macroalgae to bioenergy production in Indonesia. Thus, these assessments may provide a new perspective toward promoting and supporting the broader application of macroalgae for bioenergy production with an integrated biorefinery approach in Indonesia.

2. Macroalgae – type and characteristics

Macroalgae are divided into three groups based on pigmentation and chlorophyll content, namely red macroalgae (Rhodophyta), green macroalgae (Chlorophyta), and brown macroalgae (Ochrophyta) [35]. Red macroalgae are widely abundant species, around 4000–6000 species that grow in tropical marine environments. Meanwhile, brown macroalgae only have 1500–2000 species [36]. Brown macroalgae are found in tropical and subtropical seawater [37]. The number of green macroalgae is around 4500 species consisting of 3050 species in freshwater areas, namely *Trebouxiophyceae* classes and 1500 species that grow in marine environments for *Bryopsidophyceae*, *Dasycladophyceae*, *Siphoncladophyceae*, and *Ulvophyceae* classes [38]. In Indonesia, Santoso et al. [39] reported that macroalgae include green algae (i.e. *Caulerpa racemosa*, *Caulerpa sertularioides, Cladophoropsis vaucheriaeformis, Ulva reticulata*), brown algae (i.e. *Padina australis, Sargassum polycystum, Turbinaria conoides*), and red algae (i.e. *Kappaphycus alvarezii*), which contain high macro-mineral (i.e. Na, K, Ca, and Mg), but low trace-mineral (i.e. Cu, Zn, and Fe) contents. However, the macroalgae species in Indonesia are mainly from the genus of *Gracilaria, Eucheuma, Kappaphycus,* and *Sargassum* [40]. Our previous survey reported that several macroalgae, such as *Gracilaria verrucosa* and *Eucheuma cottonii* (Figure 1), are widely available in East Java, Indonesia.

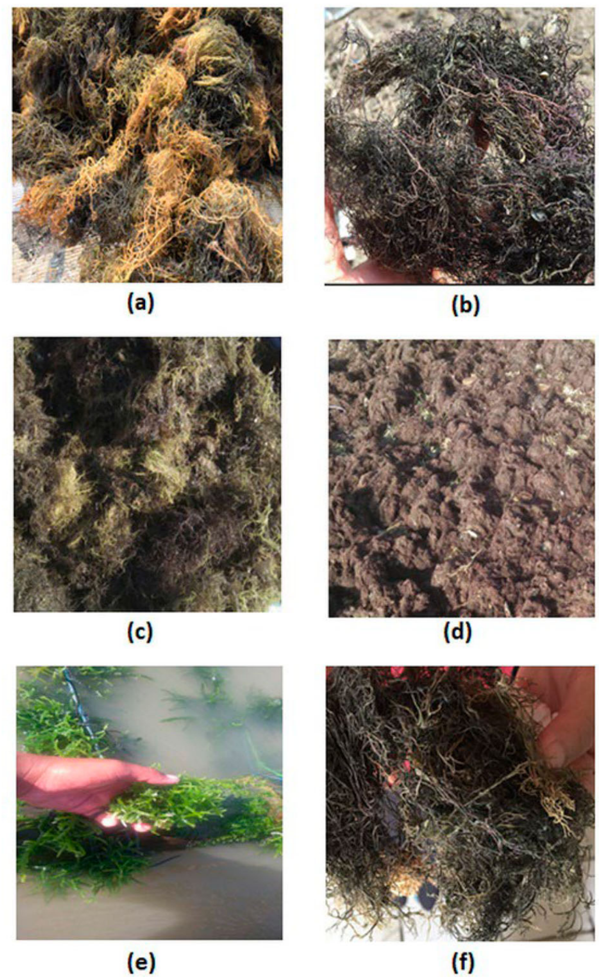

Figure 1. Images of macroalgae from East Java – Indonesia: fresh and dried cultivated *Gracilaria verrucosa* from Sidoarjo City (a and b); fresh and dried wild *Gracilaria verrucosa* from Ujungpangkah Beach (Gresik City) (c and d); fresh and dried cultivated *Eucheuma cottonii* from Sumenep City (Madura Island) (f and g). (Own photos).

The biochemical composition of macroalgae includes carbohydrates, proteins, lipids, fibre, and minerals, which differ between red, brown, or green macroalgae [41]. The main composition of macroalgae is carbohydrates, amounting to up to 50% of dry weight, lipids 1–5%, protein 10–47%, minerals 8–40%, and phenolic compounds up to 25% [42]. The biochemical compositions of various macroalgae species are presented in Table 1.

Macroalgae are suitable for biofuel production due to their high carbohydrate content [61]. However, carbohydrate content in macroalgae depends on the species, such as red macroalgae (20–60%), green macroalgae (15–50%), and brown macroalgae (10–70%). The carbohydrates in these macroalgae are produced from photosynthesis, usually consisting of monosaccharides and polysaccharides [38]. The types of monosaccharides and polysaccharides in red, green, and brown macroalgae are varied, as shown in Table 2.

The chemical compositions of macroalgae are influenced by many factors (i.e. seasonal and geographical variability, species type, water temperature, nutrients, ash content, harvest location, salinity, etc.). For instance, Adams et al. [62] revealed that the carbohydrate concentration peak of *L. digitata* was during the Summer season, which contained high mannitol and laminarin, low ash content, and low alkali index. A previous study has also highlighted the impact of seasonal variation, on the composition of brown macroalgae, with the highest carbohydrates in autumn and the lowest in winter [63]. A previous

Table 1. Biochemical composition of various type of macroalgae available worldwide and in Indonesia (on a dry basis).

Group	Species	Location	Carbohydrate (%)	Protein (%)	Lipid (%)	Refs.
Brown	*Ascophyllum nodosum*	Devon, UK	25.50	8.90	6.70	[43]
	Fucus ceranoides	Devon, UK	14.80	11.60	3.30	[43]
	Fucus vesiculosus	Devon, UK	15.90	10.50	3.80	[43]
	Himanthalia elongata	Devon, UK	23.10	9.10	2.60	[43]
	Laminaria digitata	Lysekil, Sweden	70.00	8.00	1.80	[44]
	Laminaria hyperborea	Devon, UK	17.40	13.20	2.60	[43]
	Padina boryana	Jeddah, Saudi Arabia	43.56	10.88	11.37	[45]
	Pelvetia canaliculata	Devon, UK	19.1	9.90	5.00	[43]
	Sargassum spp.	Philippines	41.81	10.25	0.75	[37]
	Saccharina Japonica	Suwon, South Korea	51	14.50	6.30	[46]
	Sargassum muticum	Devon, UK	11.30	9.90	1.50	[43]
	Sargassum plagiophyllum	East Lombok Beach, Indonesia	53.23	12.71	2.05	[47]
	Padina australis	Seribu Island, Indonesia	62.21	10.76	4.17	[48]
	Sargassum polycystum	North Borneo, Malaysia	33.49	5.40	0.29	[49]
	Turbinaria conoides	Gulf of Mannar, Tamil Nadu	14.90	15.90	3.56	[50]
Green	*Chaetomorpha antennina*	Tamil Nadu, India	15	50	2.10	[51]
	Chaetomorpha linum	Tunisia	29.76	8.60	2.60	[52]
	Cladophora glomerata	Madison, USA	25.10	29.50	8.70	[53]
	Rhizoclonium riparium	Devon, England	28.10	13.20	1.90	[43]
	Ulva sp.	Tamil Nadu, India	44.30	12.02	2.60	[54]
	Ulva intestinalis	Jeddah, Saudi Arabia	28.65	16.35	11.63	[45]
	Ulva lactuca	Southeast Maluku, Indonesia	61.83	10.00	0.13	[55]
	Ulva lactuca	Jeddah, Saudi Arabia	35.62	15.96	12.50	[45]
	Ulva linza	Jeddah, Saudi Arabia	43.60	14.27	8.30	[45]
	Ulva rigida	Sfax, Tunisia	15.88	13.69	1.55	[56]
	Caulerpa lentillifera	North Borneo, Malaysia	38.66	10.41	1.11	[49]
	Caulerpa racemosa	Southeast Maluku, Indonesia	38.62	7.60	0.71	[55]
	Ulva reticulata	Parangipettai Coast, India	0.25	19.98	1.70	[57]
Red	*Acanthophora specifera*	Jeddah, Saudi Arabia	40.22	12.58	5.43	[45]
	Chondrus crispus	Devon, UK	46.70	21.11	3.00	[43]
	Gracilaria vermiculophylla	Ria de Aveiro, Portugal	26.50–34.50	33.90–42.90	0.03–0.24	[58]
	Gracilaria multipartita	Jeddah, Saudi Arabia	50.43	14.45	5.07	[45]
	Gracilaria verrucosa (wild)	Central Java, Indonesia	60.81	9.86	0.86	[59]
	Gracilaria verrucosa (cultivated)	Central Java, Indonesia	38.38	6.64	0.58	[59]
	Gracilaria verrucosa (wild)	East Java, Indonesia	50.69	17.66	0.72	[18]
	Gracilaria gigas (wild)	Central Java, Indonesia	64.71	12.63	1.31	[59]
	Gracilaria gigas (cultivated)	Central Java, Indonesia	47.31	8.14	0.60	[59]
	Jania rubens	Jeddah, Saudi Arabia	48.76	11.85	2.77	[45]
	Laurencia obtusa	Jeddah, Saudi Arabia	41.66	10.66	4.13	[45]
	Solieria chordalis	Devon, UK	39.50	13.40	1.20	[43]
	Kappaphycus alvarezii	Various areas in Indonesia	35.60–78.30	2.30–5.40	0.40–0.90	[60]
	Eucheuma cottonii	North Borneo, Malaysia	26.49	9.76	1.10	[49]

study found that the cellulose content of brown species depended on the season and the depth of immersion where macroalgae were collected [64]. Ash and nitrogen levels also fluctuated during the year, whereas sulfur concentrations were found stable, with values typically four times lower for brown macroalgae when compared to red macroalgae. Light metals such as potassium, calcium, magnesium, or sodium were also found to fluctuate during the year. The macroalgae absorbed salts from the surrounding seawater environment, and those found in the brown macroalgae biomass include sodium, calcium, potassium, magnesium, barium, and strontium [62]. Frid [65] reported that the variability of the marine benthos might affect the functionality of the seafloor over time, allowing species fluctuations. For example, *Saccharina latissima* species are widespread on British coasts and grow in sheltered and rocky environments a little above the tidal mark until the depth of 18 m [66]. Setyawidati et al. [67] found that water motion and current significantly affect macroalgae species

Table 2. Type of carbohydrates in macroalgae.

Type of carbohydrate	Red macroalgae	Green macroalgae	Brown macroalgae
Polysaccharide	carrageenan agar cellulose lignin	mannan ulvan starch cellulose	laminarin mannitol alginate fucoidan cellulose
Monosaccharide	glucose galactose agarose	glucose manose rhamnose xylose uronic acid glucuronic acid	glucose galactose fucose xylose uronic acid Mannuronic acid guluronic acid glucuronic acid

Source: Jung et al. [38].

Table 3. Compositional analysis results of several macroalgae.

Types	MC (%WW)	Ash (%TS)	C (%TS)	H (%TS)	N (%TS)	S (%TS)	O (%TS)	CV (MJ/kg TS)	Refs.
L. digitata	3.4–6.1	13.8–34.8	26.4–36.2	4.0–5.6	1.1–3.5	0.6–0.9	30.5–42.5	10.8–14.9	[62]
L. digitata	80.3–91.6	18.3–38.3	26.1–37.2	3.4–5.5	0.9–3.95	–	21.3–39.4	–	[68]
L. digitata	6.6	25.6	35.4	5.1	1.9	0.86	36.3	14.0	[69]
L. hyperborea	5.6	17.6	37.9	5.4	1.6	0.95	41.4	15.0	
L. saccharina	6.4	23.3	34.7	4.8	1.2	0.64	40.5	13.0	
A. esculenta	6.8	27.1	37.1	5.1	2.0	0.64	33.4	14.9	
S. muticum	79.9	29.45	30.66	3.95	4.89	1.49	29.6	16.4	[70]
F. serratus	72.3	19.0	41.4	4.9	1.5	–	35.1	–	[26]
F. vesiculosus	65.9	22.3	45.1	5.1	1.5	–	36.5	–	
A. nodosum	67.4	21.2	46.4	5.2	1.5	0.7	34.8	–	
L. digitata	77.7	25.0	38.9	4.7	1.3	–	37.2	–	
L. hyperborea	73.4	16.0	42.0	5.1	0.9	–	39.0	–	
U. rigida	79.5	26.7	40.6	5.0	3.6	1.1	35.3	–	
S. latissima (wild)	74.1	31.3	40.5	4.1	1.3	0.47	45.9	9.7	[71]
S. latissima (cultivated)	82.9	33.7	35.0	3.6	1.2	0.75	36.2	11.1	
E. maxima	90.317 ± 0.09	92.472 ± 0.04	24.99	4.7	1.77	–	58.54	–	[72]
P. boergesenii	–	60.31 ± 1.90	22.82 ± 0.69	3.10 ± 0.09	1.13 ± 0.01	0.98 ± 0.01	27.14 ± 0.41	–	[73]
C. sinuosa	–	51.71 ± 1.83	24.33 ± 0.91	3.24 ± 0.21	1.62 ± 0.09	0.99 ± 0.05	27.22 ± 0.31	–	
Ulva sp.	–	40.36 ± 0.43	25.58 ± 0.46	4.61 ± 0.31	2.95 ± 0.20	3.88 ± 0.11	30.13 ± 2.52	–	
S. muticum (FD Summer)	10.9 ± 0.0	26.5 ± 0.7	33.3	5.4	3.0	0.6	30.2	–	[74]
S. muticum (Summer washed)	87.8 ± 0.1	13.6 ± 0.4	40.4	5.6	2.9	0.2	37.8	–	
S. muticum (FD Spring)	7.4 ± 0.1	26.9 ± 0.0	30.4	5.1	4.2	0.5	32.3	–	
S. muticum (FD washed)	88.8 ± 0.4	12.4 ± 0.3	41.6	6.2	3.9	0.2	35.9	–	
S. polycystum	6.7	60.2	21.8	6.2	2.5	3.6	65.9	–	[75]
G. tenuistipitata	5.9	61.2	21.6	7.0	2.1	4.1	65.2	–	
U. reticulata	11.2	47.1	24.3	6.3	2.6	6.9	59.9	–	
U. lactuca	15.10	29.21	22.99	4.55	2.44	–	–	–	[76]
L. digitata	3.5	59.9	28.6	7.8	2.6	4	57	–	[77]
A. nodosum	4.6	31.8	38.5	6.0	3.7	5.9	45.9	–	
C. linum	6.86	67.6	21.8	7.6	1.8	4.4	64.6	–	
Pelagic Sargassum	20.63 ± 0.93	31.82 ± 1.34	27.50 ± 0.65	4.16 ± 0.30	1.21 ± 0.06	0.82 ± 0.22	34.49 ± 0.18	15.66 ± 0.68	[78]
U. intestinalis	14.89	29.45	23.05	4.6	2.40	–	–	–	[79]
S. latissimi	–	24.2	33.0	2.9	1.9	ND	23.6	–	[80]
F. serratus	–	29.2	35.6	4.0	2.0	ND	27.9	–	
G. verrucosa	15.16	16.56	31.03	6.2	2.7	–	60.02	10.59	[18]
S. plagiophyllum	7.02 ± 0.02	9.32 ± 0.04	42.40 ± 0.38	5.86 ± 0.03	1.45 ± 0.01	2.78 ± 0.05	38.19 ± 0.30	14.46 ± 0.08	[47]

Note: MC = moisture content, CV = calorific value.

distribution and composition (i.e. *Sargassum*, *Padina*, and *Turbinaria*) in Libukang Island, Indonesia. Their study found that *Turbinaria* was abundant on coral reefs and soft substrates. *Sargassum* is abundant on the soft substrate but has a low probability on the hard bottom and muddy sand substrate, in contrast to *Padina*. A summary of the compositional analysis results from several macroalgae is shown in Table 3.

3. Valorisation pathways of macroalgae to bioenergy

Biofuel production from first-generation biomass (i.e. food crops) causes competition with global food security and land use. In contrast, second-generation biomass (i.e. lignocellulosic biomass) may not be favourable due to the need for pretreatment, consideration of availability and productivity land (e.g. soil infertility), depletion of freshwater supply, high energy consumption for operation, and high capital cost of conversion technology [81]. Other constraints (i.e. emission generation, low yield production, and improbability from finance) are the most common issues faced in bioenergy from lignocellulosic biomass [82]. Therefore, as third-generation biomass, macroalgae is seen as a cost-effective, fast growth rate, and sustainable feedstock for biofuel production, replacing the failure of using first – and second-generation biomass [6,20]. Over the past five years, many studies have reported macroalgae valorisation pathways (i.e. thermochemical, biological, or chemical processes) [18,20,83]. These can be classified as (1) direct combustion, pyrolysis, gasification, liquefaction, supercritical fluid extraction (thermochemical); (2) anaerobic digestion, fermentation (biological); and (3) acid hydrolysis, enzyme hydrolysis, and transesterification (chemical). The obtained biofuels for each process are categorised into bioethanol, biogas, biodiesel, bio-oil, bio-syngas, biochar, etc. (see Figure 2).

However, the valorisation pathways of macroalgae to bioenergy may also be determined by various factors (i.e. species type, sources, characteristics,

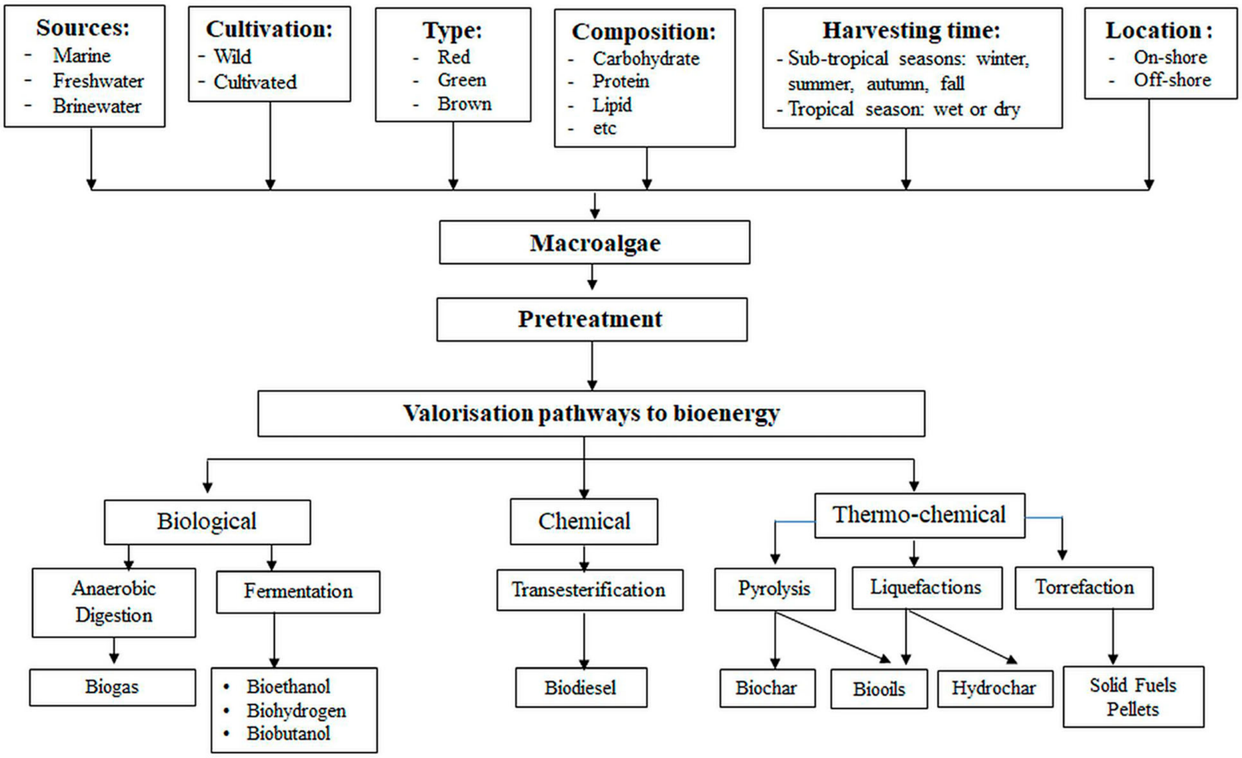

Figure 2. Valorisation pathways route for macroalgae to bioenergy.

harvesting time, location, and cultivation type). Furthermore, capturing the inherent energy content is linked to the pursued biomass conversion route. More recently, the thermochemical conversion of macroalgae biomass using direct combustion, gasification, pyrolysis, and liquefaction has been performed with positive potential [84]. Previous studies also reported that several native macroalgae in Indonesia (i.e. *Gracilaria, Eucheuma, Kappaphycus,* and *Sargassum*) could be converted to biogas [18,47] or other biofuels (i.e. bio-oil, biochar, etc.) [40]. Other studies, however, suggested that macroalgae are rich in starch and carbohydrates, therefore ideal as feedstock for bioethanol production [83,85]. Hydrothermal liquefaction is another potential method for bioenergy production from macroalgae due to its high moisture content [6,43]. Table 4 presents the summary of conversion technology for macroalgae to bioenergy with environmental, economic, and potential energy recovered.

The feasibility of using macroalgae for bioenergy production still varies, which tends to be economically unfeasible based on minimum selling price (MSP) or break event selling price (BSP). MSP value shows commercialisation potential from bioenergy products. Several studies stated that the selling price of macroalgae-based bioenergy products is still more expensive than other biomass sources. Using macroalgae as a source of biodiesel is economically unfeasible. Brigljević et al. [108,109] calculated the MSP value of macroalgae-based diesel, indicating that the process is still 1–2 times more expensive than the average price of petroleum-derived diesel. However, the economic viability of macroalgae-based bioenergy could be boosted by implementing biorefineries via the production of value-added chemicals (i.e. alginate, mannitol, carrageenan, etc.) after producing biogas, bioethanol, biocrude, or other biofuels from macroalgae. Nazemi et al. [110] found that using brown macroalgae for bioethanol (in a single production route) was still not economically feasible due to its low income or selling price. However, feasibility can be increased by combining with the production of chemicals (i.e. protein, alginate, mannitol, and fertiliser). Dickson and Liu [32] also produced an economically viable bioethanol production design by integrating succinic acid and microalgae production systems. In addition, previous research by Dickson et al. [61] showed that the MSP value of bioethanol was lower if dried distillery solids (DDS) as by-products of the bioethanol production were also sold. Such practice may increase

Table 4. Summary of conversion technology for macroalgae to bioenergy with environmental, economic and potential energy recovered.

Type of bioenergy	Conversion technology	Technology Readiness Level (TRL)	Advantages	Disadvantages	Environmental consideration	Economic consideration	Potential energy recovered	Refs.
Bioethanol	Fermentation	8–9 [86]	- Fermentation technology, either SHF or SSF, is well-established and available commercially - Combining with pretreatments (i.e. biological, chemical, mechanical, and thermal) enhanced the release of oligosaccharides and increased the bioethanol yield - Lower pretreatment temperature is required than terrestrial biomass - Higher ethanol yield compared to glucose as substrate	- Low glucose content in macroalgae leads to low bioethanol production - Inability of microorganisms to ferment all sugars present in macroalgae - Lower cellulose in macroalgae residues reduce bioethanol potential - The use of higher temperatures may result in inhibitor compounds in the fermentation process	- Macroalgal-based bioethanol has low carbon intensity (CI) and high Energy Return on Energy Invested (EROI) - Additional fermentation and distillation process may cause a higher environmental impact	- Increase cost of operation due to pretreatment requirement - The high price of macroalgal-based bioethanol	0.05–89 g ethanol/g	[37,83,87]
Biodiesel	Transesterification (with or without catalyst)	8–9 [86]	- Cut down the production cost - Produce high-quality biodiesel than diesel - The most practicable process than other thermochemical methods	- The excessive amount of alcohol will hinder the process - The lower ratio will lead to lower biodiesel yield	- Reduction of carbon emission - Non-toxic and biodegradable biodiesel - Reduction of fossil fuel consumption - Poor properties of biodiesel due to the cold weather climate	- High operational cost with more than one stage cycle - Need catalyst and reagent will impact the cost	1.24 GJ/ton lipid productivity and 1.3 GJ/ton of FAMEs	[88,89]
	Extraction – transesterification	3–8 [80]	- Simplified process with one stage - Low production cost - Reduce production time - Reduce the excessive use of solvent	- Lack of conversion efficiency compared to the conventional method - The end products have to be separated to remove impurity - High amount of reactant required - Lower reaction time - Lower FAME conversion	- Lower reagent (chemical) is needed and resulted in a greener process - Chemical co-solvent wastes and hazardous components need further treatment	- Proper handling of catalyst use will minimise the working accidents - Oil content is not as high as microalgae oil, thus lower biodiesel potential	17.1% of methyl esters yield	[90,91]
Biogas	Anaerobic digestion	8–9 [86]	- As the well-established technology, the policy	- Low gas productivity for mono-digestion	- One solution to fossil fuel shortages - Geopolitical trends	- High initial cost - Long term return on investment	0.100–0.855 m^3 CH_4/kg VS	[18,26,27,92],

			incentives are provided in many countries - Vary in scale reactor - High organic matter in macroalgae supporting the microbial activity - Fertiliser production as the by-product - Co-digestion improves methane yield	- Low substrate's degradability - Require dilution with fresh water - Need longer time to adapt to marine sediments culture - Facility corrosion in chemical pretreatment	- Seasonal harvesting affected biogas yield - AD of macroalgae has 30-fold higher environmental performance compared to others	- Some pre-treatment increases the capital cost		
Biohydrogen	– Dark fermentation – Photo fermentation	4–7 [86]	- A higher rate of H_2 production using fermentation technology - Combined (or two-stage) process provides a higher H_2 yield than single-process - Many detoxification processes are available for enhancing the H_2 production process	- Higher moisture content, ash, and alkali content hinder the H2 production - Prolong fermentation time - Need extra pretreatment to enhance H2 yield - Potential release of inhibitor as a result of pretreatment - Use of pure culture has lower organic matter degradation to H2 - Commercial application of the technology is currently not applicable	- Low greenhouse gas (GHG) emission - Biohydrogen is considered as a clean fuel - Some pollutants were generated alongside the process - Catalyst use with green chemistry consideration	- Addition of pretreatment and detoxification steps increases operational cost - Commercial application of technology is not available yet - Finding a low-cost catalyst to balance the cost	28–102.7 mL H_2/ g dry algae	[19,93,94]
Biochar Bio-oils	Pyrolysis	5–8 [95]	- Readily commercial with lower cost (in the stable process) - Pyrolysis technology is highly flexible for any biomass - No chemicals requirement - Pyrolysis of macroalgae requires less energy and low temperature than woody biomass - A lower phenolic compound in macroalgae than woody biomass leads to a better deoxygenation process - Macroalgae have a higher bio-oil yield (65% ww) than lignin (47% ww)	- More space for the location of the plant - Ex-situ configuration may result in char and gas production - Safety issues dealing with the use of H2 at a higher temperature - Complex operation - Resulted bio-oil may contain impurities - Only could maintain some algae due to the limitation of moisture content - Coke formation caused the loss of catalytic activity	- Lower CO_2 emissions - Reusability in catalyst enhance the sustainability principle - Heavy components from the process need further improvement	- High capital cost - High operational cost from the use of catalysis and maintenance cost - Additional economic benefits from the use of biochar as coil enhancement or fertiliser	Bio-oil yield: 11.0–47.4% Biochar yield: 30–50.35%	[96–99]

(*Continued*)

Table 4. Continued.

Type of bioenergy	Conversion technology	Technology Readiness Level (TRL)	Advantages	Disadvantages	Environmental consideration	Economic consideration	Potential energy recovered	Refs.
	Hydrothermal Liquefaction (HTL)	3–8 [80]	- Applicable to a wide variety of biomass type - High quality of bio-oil than the pyrolysis - Skip the need for the dewatering - Reduce the issues about fouling and slagging	- Require high temperature and pressure - Viscous - Unflavoured energy balance - Need to remove the salt content in the initial process - Massive use of the water - High energy loss	- More handling the water as process water through recycling and reuse concept - Prevent the discharge of water directly	- Low capital cost (no demand to involve the dewatering) - Energy-saving from the reduced oxygen - Low economic feasibility (process water use)	Bio-oil yield: 20–63%	[96,100,101]
	Gasification	4–7 [102]	- The outcome can be used either in generating electricity or chemical products - High conversion efficiency - Low reaction time - Combination technology will enhance the quality of biofuel	- Low calorific value gas - Formation of tar or hydrocarbon condensate hinders the process - The corrosion and fouling issues - Low calorific value gas - Energy loss due to the heat exchanger limitation - Need salt removal before entering the gasifier	- Different cultivate locations will show distinct properties of algae (ash and alkali metal concentration) - Require additional water for dilution	- High operational cost due to the obligation of drying method - Operational cost tends to be high for water use		[96,103,104]
Biobutanol	Aceton-Butanol-Ethanol (ABE) fermentation	8–9 [105]	- Easily seeking investment - High energy density - Less corrosive - Low hygroscopic	- High use of energy and water - Low butanol yield - Low ABE's density - Significant production of by-products and butanol toxicity - Need more intensive maintenance during running production - Must provide a high load to meet the optimum result	- Poor emission performance (under non-optimum conditions) - A solution to the depletion of fossil fuel resources	- High capital cost - High operational cost	93.4 mg ABE/g algae	[101,106,107]

the macroalgae's economic feasibility as a source of thermochemically produced bioenergy. Greene et al. [111] concluded that the MSP value for diesel is cheaper by including by-product revenue in the form of nitrogen and phosphorus fertilisers. Economic feasibility can also be increased by co-substrate. For instance, Abohmora et al. [45] produced a lower estimated selling price of bio-oil from macroalgae co-substrates with low-density polyethylene (LDPE) plastic than bio-oil and biodiesel on the market.

3.1. Bioethanol

Bioethanol is a liquid biofuel that is produced from microbial degradation of any sugar-containing materials; the process consists of several stages, including pretreatment, hydrolysis (i.e. enzymatic or acid), filtration, fermentation, and purification(i.e. distillation) [112]. Bioethanol production from first – to second-generation biomass substrates has been widely investigated. However, it has shown inadequate performance due to competition with food crops, land use issues, and a highly cost – extensive delignification [83]. Therefore, bioethanol production from third-generation biomass (i.e. algal biomass) is currently seen as an attractive solution due to its high yield compared to terrestrial plants [101]; no competition with land use for food supply, can be grown in wastewater and fresh water, as well as accumulate large quantities of carbohydrates and lipids [83]. Furthermore, compared to terrestrial bioethanol plants, macroalgae contain a higher level of polysaccharides and sugar alcohols, lower lignin and cellulose content, as well as similar bioethanol potential (i.e. 29.6 kg bioethanol/tons wet macroalgae), making it a potential candidate for feedstock [6].

Various studies have emphasised the potential conversion of macroalgae into bioethanol, with and without pretreatment. For example, Adams et al. [113] studied bioethanol production from *Laminaria digitata* before and after washing pretreatment (for 1 min), followed by drying (for 72 hrs at 70°C) and milling to obtain the particle size of < 1 mm. Their results suggested that unwashed *L. digitata* produces bioethanol more efficiently than washed *L. digitata.* This was believed to be due to a decrease in water-soluble carbohydrates. Kostas et al. [114] studied a laboratory-scale experiment that utilised a yeast strain following an acid pretreatment to improve bioethanol production from *Laminaria digitata*, *Fucus serratus*, *Chondrus crispus*, *Palmaria palmata,* and *Ulva lactuca*. The results suggest that using *Saccharomyces spp* resulted in the highest ethanol production from the hydrolysates of *P. palmata* and *U. lactuca*, with a value of 7 g/L. Their study also reported that the macroalgae samples have the potential valorisation for xylitol, succinic acid, and acetic acid production. In Indonesia, Poespowati et al. [115] studied the bioethanol production from *Ulva lactuca* locally found on southern coast of Central Java utilising different chemical pretreatments, including citrate and acetic buffer. Before chemical pretreatment, macroalgae samples were washed, sun-dried (for 4 days), and grounded (80 *mesh*) to improve the process efficacy. The results showed that within 10 days of fermentation of citrate buffer treated macroalgae, an optimum ethanol yield was obtained, counted for 0.985% (or 9.85 $g/kg_{macroalgae}$). Other studies have reported the bioethanol potential from different types of macroalgae found in Indonesia, such as *Eucheuma cottonii* [116], *Palmaria palmata* [117], Sargassum crassifolium [118], as well as *Gracilaria gigas* and *Gracilaria verrucosa* (wild and cultivated) [59].

There are various challenges in producing bioethanol from macroalgae, especially in improving the hydrolysis and fermentation efficacy. The expensive cost for an enzyme to break down the alginate content in macroalgae remains a critical problem, where using modified microorganisms can enhance the fermentation of alginate [6,94]. Enquist-Newman et al. [119] also suggested that major engineering modification is required to enable *Saccharomyces cerevisiae* effectively transform brown macroalgae sugars into bioethanol.

3.2. Biodiesel

Biodiesel, as one alternative liquid biofuel with lower emission than diesel, can be produced either by conventional transesterification process with and without catalytic [120], ultrasound assisted-transesterification [121], or by extraction of fats/oils followed by fermentation [122]. To generate biodiesel, substrates with high lipids/fats or oil content is critical to ensure a better process efficacy and high yield [123]. Producing biodiesel from macroalgae consists of several stages, including oil extraction, transesterification, and alcoholysis [124]. Mechanical pretreatments are often used to prepare the macroalgae substrates, including washing, drying, and milling [91]. After mechanical pretreatment, macroalgae was subjected to oil extraction steps (mostly using chemical methods). Then, in the transesterification step, the macroalgae oil is converted into biodiesel, where a triglyceride will react with alcohol. The glycerol produced during the process can be further valorised in the food and cosmetic industry [91,124]. However, several studies have pointed out that dried macroalgae can be directly covert into biodiesel without an oil extraction step (i.e. direct transesterification). This may significantly reduce the processing time and the operational cost (i.e. chemical solvents and energy) [20,90].

Algal oil has been highlighted as feedstock for economically and eco-friendly biodiesel production [91]. Several studies have explored the conversion of macroalgae to biodiesel. For instance, brown macroalgae (*P. tetrastromatica*) was an attractive feedstock to produce high-quality biodiesel and meet the standard required (STM D6751) [125]. Three marine macroalgae (i.e. *Ulva* lactuca, *Padina boryana,* and *Ulva intestinalis*), due to their high lipids and fatty acid methyl esters (FAMEs) contents, were found to produce high-quality biodiesel, following that of the international standard [67]. Their study also confirmed that the solid residual fraction from biodiesel extraction was potential for biogas production as it still contains high fermentable sugar with less oil content. Similarly, Saengsawang et al. [91] reported that *Rhizoclonium* sp pre-treated with ultrasonication enhances the oil yield extracted, thus improving the quality and quantity of the resultant biodiesel. Maceiras et al. [126] investigated the conversion of various macroalgae (i.e. *Fucus spiralis, Saccorhiza polyschides*, *Sargassum muticum, Codium tomentosum, Ulva rigida, Enteromorpha intestinalis*, *Ascophyllum nodosum*, *Pelvetia canaliculata*, *Heterosiphonia plumose*, *Calliblepharis ciliate*, *Ceramium rubrum*, *Polysiphonia lanosa, Bifurcaria bifurcate,* and *Plocamium cartilagineum*) into biodiesel via extractive-transesterification, which showed a positive result. The oil content of the macroalgae species varied from 20 to 50%, with 1.65–5.14% conversion to biodiesel.

Various problems in converting macroalgae to biodiesel, such as transesterification via extraction method, require high amount of solvent and lower FAME conversion and reaction time [104]. Chen et al. [22] suggested that transforming macroalgae into biodiesel is not the best option due to its lack of triglycerides content. Yet, biodiesel's technology advancement and process efficacy from macroalgae still need to be further investigated.

3.3. Biogas

Anaerobic digestion (AD) has been highlighted as a sustainable technology for converting macroalgae into biogas, a mixture of methane, carbon dioxide, and other trace gases. In principle, AD that runs simultaneously without the presence of oxygen in four steps (i.e. hydrolysis, acidogenesis, acetogenesis, methanogenesis) is part of the biochemical process to convert the organic substrates under the optimum conditions (i.e. pH, temperature, salinity, nutrients) into methane as end-product. The condition of substrates as the AD's feedstock is the most crucial variable to determine the performance of AD itself, besides other factors (e.g. inoculum, digestion test, and digestion system) [127]. Pre-treatment is necessary to breakdown the recalcitrant of the cell wall and enhance the biodegradability of substrates. Whether physical, chemical, biological, or combination of those, the choice of selection is based on substrate's characteristics and feasibility of operation [128].

The AD of macroalgae has been demonstrated with high gas yields and conversion rates. Numerous studies have investigated AD of macroalgae either in a mono – or co-digestion system, as shown in Table 5. For instance, Hinks et al. [129] investigated biogas production from *L. hyperborea* using batch and continuous systems. Their studies reported that AD resulted in biogas with a 60–70% methane composition, with specific methane potential (SMP) values of 0.250–0.410 m^3/kg TS. Furthermore, Tedesco and Stokes [26] investigated the valorisation of residues of six indigenous Irish macroalgae species (i.e. *Fucus serratus, Fucus vesiculosus, Ascophyllum nodosum, Laminaria digitata, Laminaria hyperborean,* and *Ulva rigida*). The macroalgae residues were obtained from the extraction of alginic acid, fucoidan, fucoxanthin, laminarin, mannitol, and proteins following the biorefinery. The biochemical methane potential (BMP) test was employed in this study. The results showed that the residues from *Laminaria* and *Fucus spp.*

Table 5. Studies on macroalgae for biogas production.

Species	Pretreatment	Operational condition	SMP (m^3/ kg VS)	Ref.
Laminaria sp.	A Hollander beater (MG and BT factors)	The inoculum was not degassed, HRT 21 days, batch operation	0.084–0.391	[139,140]
L. digitata	Washed and oven-dried	Each reactor filled up with 2.5 g VS/L of substrate and 4 g VS/L of sludge (S:I of 1:1.6) at 35°C, HRT 35 days	0.203	[113]
	Unwashed and oven-dried		0.235	
	Washed, frozen, and oven-dried		0.248	
	Unwashed, frozen, and oven-dried		0.192	
	Washed and freeze-dried		0.258	
	Unwashed and freeze-dried		0.240	
Ulva lactuca	No pretreatment	Six continuously stirred-tank reactors (CSTR) in 5 L of volume operated for 42 weeks at 37°C, initial organic loading rate (OLR) of 2 kg VS m^3/day	0.174–0.177 (CSTR) 0.183–0.210 (BMP)	[141]
Laminaria sp.	Harvesting time and beating	The inoculum was not degassed, batch mode at 38°C, HRT 25 days, I:S ratio of 6:8 (on a VS basis)	0.342	[142]
Laminaria sp	Untreated	The inoculum was not degassed, batch system, various S:I ratio (0.34, 0.83, and 1.34), initial pH at 7 and 8, HRT 14 days	0.328	[27]
	Beating (BT)		0.335	
	Ball milling (BM) 1 mm		0.241	
	Ball milling (BM) 2 mm		0.260	
	Microwave (MW)		0.244	
L. digitata	Co-digestion with green peas (2–35% TS macroalgae addition)	A single-stage reactor in initial batch mode at 37°C, HRT 17 days	0.275–0.325	[143]
L. digitata	Seasonal variation	Batch mode at 37°C, continuously mixed at 45 rpm speed, I:S ratio of 1:2 (on a VS basis)	0.203–0.327	[68]
L. digitata	Maceration after hot washing	Batch mode at 37°C, continuously mixed at 30 rpm speed, I:S ratio of 1:2 (on a VS basis)	0.282	[144]
F. serratus	No pre-treatment. Samples were residues after extraction	Batch mode at 39°C, pH ranged from 8.9–9.3, Incubation time of 15 days, S:I ratio of 1:3	0.101	[26]
F. vesiculosus			0.103	
A. nodosum			0.084	
L. digitata			0.187	
L. hyperborea			0.195	
U. rigida			0.073	
S. lattisima (wild)	Maceration	S:I ratio of 1:6, batch mode at 37°C with stirring for 30 days	0.249	[145]
S. lattisima (cultivated)	Maceration	S:I ratio of 1:6, batch mode at 37°C with stirring for 30 days	0.393	
G. verrucosa	Unwashed	S:I ratio of 1:6, batch mode at 37°C without shaking for 28 days	0.018	[146]
	Washed		0.109	
G. verrucosa (wild)	Unwashed and grinded	S:I ratio of 1:6, batch mode at 37°C without shaking for 28 days	0.069	[18]
	Washed and grinded		0.092	
	Unwashed, mono-and co-digestion with tofu dregs		0.045–0.216 (depend on substrate ratio)	

Note: OLR = Organic loading rate; SMP = Specific Methane Production.

produced higher methane at values of 0.187–0.195 m^3 CH_4/kg VS and ~0.100 m^3 CH_4/kg VS, close to values for raw un-extracted macroalgae. Sitompul et al. [130] studied *Ulva lactuca* from Indonesia as a potential feedstock to be valorised for biogas production. Their research suggested that *U. lactuca* can produce approximately 4.88 L of biogas with 49.90% methane concentration. Kawaroe et al. [131] reported that *Eucheuma cottonii* has a biogas potential of 4.16 L/day. A study on AD of *Gracilaria* sp under batch conditions by Kawaroe et al. [132] has also reported that these macroalgae are a potential feedstock for biomethane production, with a value of 11.6 l CH_4 /kg. Krisye et al. [133] found that *Ulva* sp. has a biogas potential with a production volume of 70.9 L from 8.8 L *Ulva* sp. Oktiana et al. [134] found that a continuous AD of *Ulva* sp. (non-edible macroalgae) can generate biogas and methane potential of 0.770 m^3 biogas/kg COD and 0.330 m^3CH_4/kg COD, respectively. The various species of macroalgae exhibit different biogas and methane potential. Our previous studies have also highlighted that *Gracilaria verrucossa* has potential as anaerobic mono – and co-digestion feedstock, with maceration and washing pretreatment, enhancing the SMP values [18,135]. Therefore, compared with other biomasses (i.e. agricultural crop residues and agro-industrial waste), the methane potential from macroalgae is within the range. For example, Brulé et al. [136] reported that methane potential from energy crops (i.e. green cuttings and grass silage) was in the range of ~0.300–0.350 m^3/kg VS. Suhartini et al. [137] found that the methane potential of sugar beet pulp was in the range of 0.280–0.360 m^3/kg VS (under mesophilic or thermophilic condition). Suhartini et al. [138] also reported the variation of methane potential of agricultural residues (0.181–0.420 m^3CH_4/kg VS) and agro-industrial waste (0.262–0.407 m^3CH_4/kg VS), depending on the waste composition.

However, in AD of macroalgae, various challenges need to be tackled, such as high nitrogen content in macroalgae causing a lower C/N ratio of ~10 (making it non-ideal for AD process), high ammonia nitrogen (causing toxic conditions to methanogens bacteria), and considerably high lignin content (making it hard to access the organic matter) [96]. Furthermore, various factors or parameters contributed to the discrepancies in methane yield of the several studied algae species, including pre-treatment, seasonal variations, composition (i.e. total nitrogen, cellulose, hemicellulose, and lignin, macro – and micronutrients presence, etc.), and addition of co-substrates (known as co-digestion) [47,63,73].

3.4. Biohydrogen

Macroalgae is one of the suitable substrates for biohydrogen production due to the high availability of carbohydrates and lipids with low lignin [147]. As a sustainable and clean energy, biohydrogen is mainly generated from the anaerobic fermentation processes (i.e. dark and photo fermentation) and bio-electrochemical processes (i.e. microbial electrolysis cell and fuel cell). The anaerobic fermentation method has advantages over other methods, such as its higher feasibility and viability [148]. Dark and photo fermentation require less energy input and zero pollution emitted. Among the mentioned technologies, dark fermentation is widely applied due to its lower cost, energy consumption, and fast rate of biohydrogen conversion [149]. Besides the advantages, the tightly bonded macroalgae cell wall limits the conversion process. Hence, some strategies are being conducted (e.g. pretreatment, fermentation system, etc.). A study by Ding et al. [150] improved the two-stage fermentation by combining with various pretreatments to depolymerise *Laminaria digitata* for enhancing biohydrogen and biomethane yield. The optimum pretreatment was hydrothermal pretreatment (HTP) at 140°*C* for 20 min, producing biohydrogen and biomethane at 44.8 and 282.2 mL/gVS, respectively. The total energy yield (10.6 kJ/gVS) was 26.7% higher than untreated macroalgae. Previously, Lin et al. [93] also improved the two-stage fermentation of *Saccharina latissima* and assessed HTP (140°*C* for 30 min) as the optimum pretreatment. The yield was 10.7 L/kgVS of biohydrogen and 345.1 L/kgVS of biomethane, giving a higher energy yield of 12.5 MJ/kgVS (or 22.5% higher) than untreated *S. latissima*.

The utilisation of macroalgae co-substrate with other biomasses has also been investigated. Ding et al. [151] carried out two-stage batch-fermentation of co-substrate of macroalgae (i.e. *Laminaria digitata* and *Saccharina latissima*) and microalgae (i.e. *Chlorella pyrenoidosa* and *Nannochloropis oceanica*). This study exhibited that *L. digitata* as a mono-substrate can produce a higher biohydrogen of 81.8 mL/gVS than other mono-substrates (after 4-day fermentation) due to the C/N ratio (28.3). However, co-substrate *L. digitata* with *C. pyrenoidosa* enhanced hydrogen yield up to 97.0 mL/gVS with a C/N ratio of 20. The methane yield was 224.3–295.9 mL/gVS after operating for 26 days. Its study also marked that the energy conversion efficiency (ECE) of first-stage dark fermentation was 4.6–6.6% and increased to 57.0–70.9% in the two-stage fermentation. Then, Ding et al. [152] conducted a study on two-stage continuous-fermentation of co-substrate of macroalgae (*L. digitata*) and microalgae (*Arthrospira platensis*). It obtained a specific hydrogen yield of 55.3 L/kgVS after 4 days with an OLR of 6.0 gVS/L/day, and the highest specific methane yield was 245.0 mL/gVS during 12 days of hydraulic retention time (HRT) with an OLR of 2.0 gVS/L/day. The energy yield was 9.4 MJ/kgVS. This result was lower than the two-stage batch fermentation system. However, the hydrogen potential of macroalgae *L. digitata* is slightly higher than sugar beet pulp, which produces hydrogen of 37.16 L/kgVS with an OLR value of 8.02 gVS/m^3/d for 5 days. The fruit and vegetable waste feeding at OLR 15 gVS/m^3/d for 3 days obtained hydrogen at 52.1 L/kgVS, then corn silage was moderate, reaching only 26.63 L/kgVS [153].

3.5. Biochar

Under high temperatures ranging from 300° to 700°C with limited (nearly zero) oxygen supply or pyrolytic conversion, macroalgae could be processed into bioactive compounds and biochar, which are beneficial for biofuel, waste treatment, and soil amendment. Given a suitable composition and structure, macroalgae-based biochar could be an option for eco-friendly and cost-effective material [154]. Macroalgae-based biochar has a high specific surface area, pore volume and distribution, sorption capacity [155], and a lower C/N ratio making it useful for direct nutrition [156].

Previous research from Kebelmann et al. [157] found that intermediate pyrolysis technology can transform macroalgae into bioenergy and biochemicals. Their study revealed that Polar macroalgae from the Arctic region Kongsfjorden and the Antarctic peninsula, Potter Cove King George Island (such as *Prasiola crispa*, *Monostroma arcticum*, *Polysiphonia arctica*, *Devaleraea ramentacea*, *Odonthalia dentata*, *Phycodrys rubens*, *Sphacelaria plumose, Gigartina skottsbergii*, *Plocamium cartilagineum*, *Myriogramme manginii*, *Hymencladiopsis crustigena*, and *Kallymenia Antarctica*) have approximately 33–46% biochar residues and 27–45% ash contents. Milledge et al. [70] reported that *Sargassum muticum* could be valorised as a bioenergy source with biochar production (67.6%TS) using slow pyrolysis technology. Similarly, another study reported that using a slow pyrolysis process at 450°C for 60 min, the selected macroalgae produced between 45 and 62% biochar, with no significant difference between species [40]. Their study has also revealed that combining biochar from macroalgae and lignocellulosic biomass can improve crop productivity due to the high fixed carbon and mineral-rich content in the biochar produced. Slow or conventional pyrolysis produces more charcoal (35–40% by weight), whereas around 60–80% by weight bio-oil is produced by fast pyrolysis. Due to their unique mineral characteristics, biochars can be used for soil amelioration. The yield and elemental analysis results from various macroalgae found in Indonesia are seen in Table 6.

3.6. Bio-oil

Liquid product, bio-oil, extracted from macroalgae through hydrothermal liquefaction (HTL) has shown that the process does not require high energy and operational cost [158]. HTL is a biochemical process that uses a specific temperature (>200°C) and pressure (>5 MPa), with or without chemical addition. A high yield and high bio-oil quality will be achieved under the optimum conditions through the process parameters (i.e. substrate composition, catalyst use, co-solvents, pH, temperature, and pressure) [159]. Research from Kostas et al. [160] investigated *Laminaria digitata* and extraction residue of *Laminaria digitata* from bio-oil production via microwave pyrolysis. The biocrude yields were obtained at around 5–8%, with residues of10–14%, respectively. However, the microwave pyrolysis required an average energy of 1.84–2.83 kJ/g and obtained unique products such as L-Proline, 1-methyl-5-oxo-, and methyl ester. Raikova et al. [43] also used HTL technology on various macroalgae species (i.e. *Ascophyllum nodosum*, *Chondrus crispus*, *Fucus ceranoides*, *Fucus vesiculosus*, *Himanthalia elongata*, *Laminaria digitata*, *Laminaria hyperborea*, *Pelvetia canaliculata*, *Rhizoclonium riparium*, *Sargassum muticum*, *Solieria chordalis*, *Ulva intestinalis,* and *Ulva lactuca*) to produce bio-oil, biochar, gas, and aqueous phase products. The bio-oil yields were approximately 6–30%, with the higher heating value (HHV) ranging from 28.4 to 33.0 MJ/kg, and the highest yield was 29.9% from *Ulva lactuca*. While the biochar yields were around 23–51%, with the highest of *Chondrus crispus*. The enhancement of bio-oil yield decreases in a solid phase (biochar). Compared to other biomass, the

Table 6. Compositional analysis results of biochars from several macroalgae commmonly found in Indonesia [40].

Types	Yield (%WW)	C (%TS)	H (%TS)	N (%TS)	S (%TS)	O (%TS)	CV (MJ/kg TS)	Operational Condition
Gracilaria (South Sulawesi)	59.8	30.9	2.2	2.8	4.4	16.5	16.1	Milled powder macroalgae processed at slow pyrolysis
Gracilaria (Java)	61.8	24.5	1.5	1.3	2.7	19.8	11.1	
Euchuema (South Sulawesi)	61.7	25.6	1.8	0.8	9.3	24.9	17.2	
Euchuema (Java)	57.2	23.7	1.2	0.7	7.0	20.6	14.6	
Kappaphycus (South Sulawesi)	59.2	31.3	2.1	0.7	6.8	23.8	17.8	
Sargassum	49.0	29.1	2.0	1.0	0.9	15.3	11.8	

biocrude yield of aspen wood was 17.0–70.3% wt and 37.4 MJ/kg of HHV [161]. While the sewage sludge had 39.46 ± 1.16% wt with 36.14 MJ/kg of HHV, *Spirulina* had 34.51 ± 1.31% wt with 34.33 MJ/kg of HHV, and rice straw had 21.14 ± 0.93% wt with 33.90 MJ/kg of HHV [162]. A previous study by Smith and Ross [163] found that *L. digitata*, *L. hyperborean,* and *A. esculenta* provide biocrude yields of 18.4–21.8%, 23.6–39.0%, and 23.7–30.0%, respectively.

A study by Anastasakis and Ross [69] found that HTL technology effectively produced bio-oils. In this study, four brown kelps collected from the west coast of Scotland (i.e. *Laminaria digitata*, *Laminaria saccharina, Laminaria hyperborean*, and *Alaria esculenta*) were valorised into bio-oils (in the range of 9.8–17.8% wt) and bio-chars (in the range of 10.9–18.6%wt). Valorisation using HTL technology demonstrated that both *L. saccharina* and *A. esculenta* have higher energy balance than other species, 7.91 MJ/$kg_{macroalgae}$ (HTL) and 8.25 MJ/$kg_{macroalgae}$ (AD), respectively. The energy potential was similar to that of anaerobic digestion (AD) technology. This aligns with Anastasakis and Ross [164], who reported that the aqueous phase from the HTL technology was rich in sugar, ammonium, potassium, and sodium, offering the potential valorisation to other high-value-added products. Meanwhile, Milledge et al. [70] reported that *Sargassum muticum* treated with slow pyrolysis produced bio-oils and biochar.

3.7. Biobutanol

Since biobutanol has become a promising renewable liquid fuel due to its high similarity with gasoline, many researchers have been attracted to find suitable feedstock and processes in biobutanol production, including macroalgae biomass. Despite containing rich carbohydrates, macroalgae must be hydrolyzed to form fermentable sugars. A pretreatment could disrupt the macroalgae's cell wall, providing the easily degradable materials in the hydrolysis step. Chinwatpaiboon et al. [165] observed a positive change in the hydrolysis efficiency and total sugar recovery via alkaline pretreatment on aquatic biomass. In contrast, López-Linares et al. [166] found that applying hydrothermal pretreatment has no significant in improving sugar recovery and yield results. Hence, the chosen pretreatment method should be considered based on the substrate's compounds and other factors (i.e. feasibility, sustainability). After the pretreatment, andenzymatic hydrolysis, an acetone-butanol-ethanol (ABE) fermentation by microorganisms will take place.

To date, only a few laboratory-scale studies that reported macroalgae-based biobutanol production, mainly in developed countries. For example, Bikker et al. [167] carried out a study of *Ulva lactuca* from the Irish coast for biofuels production through ABE fermentation by *Clostridium beijerinckii* using the hydrolysate (i.e. glucose, rhamnose, and xylose) as substrates. Their study found butanol as the primary end-product of the ABE process in high yield (5–8.5 g/L butanol product or ~0.17–0.20 g butanol/g total sugars). In the previous research, Van der Wal et al. [168] also studied *Ulva lactuca* from the North Sea (Dutch coast), obtained butanol in the range of 6.8–10.7 g/L or yielding ~0.23 g/g total sugars by *C. beijerinckii*. Moreover, this study also obtained a butanol yield of ~0.17 g/g total sugars by *C. acetobutylicum* ATCC 824. While Hou et al. [169] fermented *Laminaria digitata* hydrolysate from the North Sea (Danish Coast) by *Clostridium beijerinckii* DSM 6422 to produce butanol via ABE fermentation. This study reached a high yield of 0.27 g/g total sugars. Compared to other biomasses, the butanol yield of macroalgae was within the range; some studies even reported higher values compared to agricultural crops or residues. For example, potato peel can produce a butanol yield of 0.1 g/g total sugars by *C. beijerinckii* and 0.01–0.02 g/g total sugars by *C. acetobutylicum* [170]. The butanol yield of Eucalyptus sawdust is 0.15 g/g total sugars by *C. beijerinckii* DSM 6422 [171]. Md Razali et al. [172] revealed that oil palm empty fruit bunch has a butanol yield of 0.16 g/g total sugars by *C. acetobutylicum* ATCC 824, while Guan et al. [173] obtained a yield of 0.18 g/g total sugars by *C. acetobutylicum* from paper sludge substrate. The findings indicate that hydrolysate macroalgae can be valorised for butanol production by *Clostridium sp*, yet further improvement and feasibility study are required for scale-up and commercialisation. Nonetheless, Green Biologics (UK), the biobutanol industrial global leader of renewable sources, has set guidelines to produce biobutanol on a large scale from macroalgae feedstock [174]. However, there has been limited information available on biobutanol production using macroalgae in Indonesia.

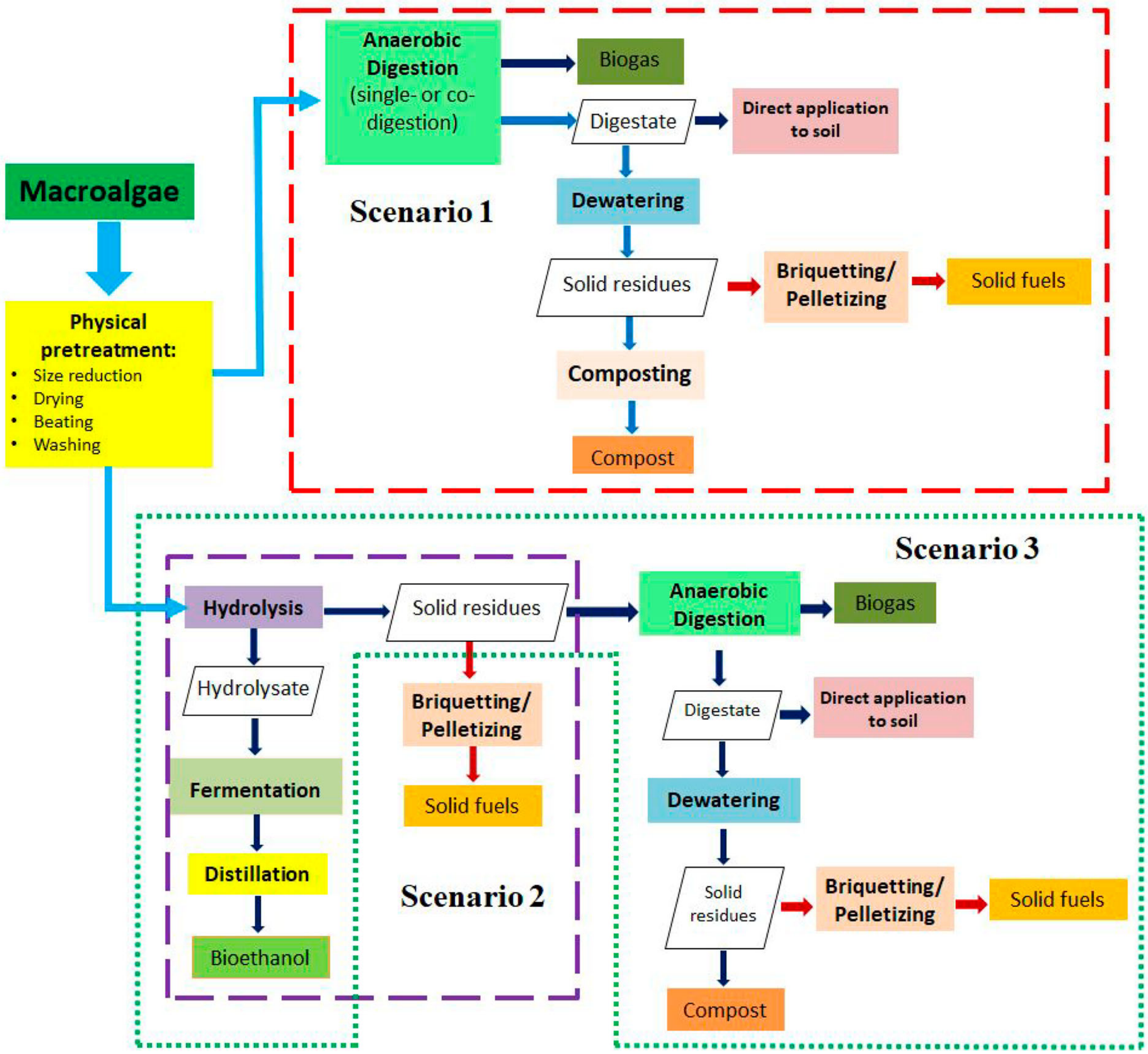

Figure 3. Proposed biorefining of macroalgae in Indonesia.

4. Biorefining of macroalgae for bioenergy in Indonesia

Based on the review, the proposed biorefining of macroalgae that may provide better economic and environmental benefits in Indonesia is AD technology and bioethanol fermentation, or a combined system with integration of composting, briquetting, or pelletising unit, as shown in Figure 3. Three scenarios of macroalgae valorisation to bioenergy could be applied. First is Scenario 1, which uses AD technology as the primary process to produce biogas and digestate. The biogas is used for cooking, replacing liquefied petroleum gas (LPG), or converting it into electricity. While the digestate is used as follows: (1) a whole digestate biofertilizer was directly applied to soil or (2) subjected to dewatering which generated liquid fraction (can be used for liquid fertiliser) and solid residues. The solid residues can be composted to produce compost or densified using briquetting or pelletising technology to generate solid fuels. Second is Scenario 2, consisting of bioethanol production as the main process, followed by briquetting or pelletising for solid residue recovery. Finally, Scenario 3 combines bioethanol production as the main process with AD system for treating the solid residues from bioethanol extraction to generate biogas, compost, or solid fuels as co-products.

Biorefining macroalgae for biogas production with a combination of compost and solid fuels production, as in Scenario 1, must consider the various parameters' impact on the AD's performance and efficacy. These include the need for pretreatment, anaerobic co-digestion, and the effect of season variability and chemical composition.

4.1. Impact of pretreatment

This section further explores the effect of pretreatment on biogas or methane production from macroalgae, as shown in Table 5. Many studies have found some findings to support its application for better macroalgae AD performance. Tedesco et al. [139,140] studied the pretreatment of particle size reduction to improve the biogas or methane production from *Laminaria* sp., collected from Ireland. A Hollander beater was used to reduce the particle size of *Laminaria* sp., combining the factor of machine gap/MG (i.e. 0, 5, and 10 turns) and beating time/BT (i.e. 5, 10, and 15 min). The results illustrated that pretreatment of *Laminaria* sp. resulted in higher biogas production ranging from 0.201 to 0.978 m^3/ kg VS and methane in 0.084–0.391 m^3/kg VS, respectively. The findings further confirmed that reducing the particle size of *Laminaria* sp. to below 1.6 mm enhanced biogas and methane production by up to 52–53%. As part of their studies, Adams et al. [113] also investigated the impact of washing as a pretreatment on biogas production. The results found that unwashed *L. digitata* generated lower biogas than washed *L. digitata* due to the presence of salts (e.g. sodium), which contribute to a decrease in the growth of microbial consortia within the AD. Our previous studies also demonstrated that washing pretreatment on *Gracilaria verrucosa* improves SMP value by 5-fold from 0.018–0.109 m^3 CH_4/kg VS [146], while co-digestion improved methane from 0.045–0.120 m^3 CH_4/kg VS [18]. Allen et al. [175] studied ten macroalgae species found around the Ireland coastline as a potential resource for methane production. The results showed that the methane potential ranged from 13.5 to 34.5 m^3 CH_4/tons WW. Compared to other species, *S. latissima* produced the highest methane potential with a value of 0.342 m^3 CH_4/kg VS. Their calculation further showed that cultivation of *S. latissima* is recommended, which may produce approximately 0.250 m^3 CH_4/ha/year, exceeding that of the land biomass. Montingelli et al. [27] compared treated and untreated *Laminaria* sp for biogas production using AD technology (continuous stirred reactor tank/CSTR). The results indicated that *Laminaria* sp., which was treated with a beating pre-treatment of 5 min, gave the highest methane yield of 0.342 ± 0.017 m^3/kg VS due to the higher value of TS (19± 2% WW) and VS (84 ± 1% TS) than two other feedstocks. These findings showed that *Laminaria* sp. is a suitable feedstock for AD, and biogas and methane yields can be optimised with a combination of physical pretreatment. Tabassum et al. [144] investigated the impact of hot washing (40 °C) prior to maceration on the methane production from *L. digitata*. Their study found that the pretreatment could improve the VS content by 31%, reduce salt accumulation by 54%, and increase biodegradability by 16%. Such improvement thus led to a 25% increase in methane yield counted at 0.282 m^3/kg VS. The above studies indicate that macroalgae can be valorised into renewable energy with and without pretreatment. Yet, the pretreatment itself has been found to increase the efficiency of the conversion process. Further investigations into the impact of pretreatment on the operating cost and energy requirement are needed to evaluate the commercial viability of the conversion pathways.

4.2. Impact of seasonal variation

A previous study by Paul et al. [176] also confirmed that AD effectively valorises macroalgae. In this study, the role of pretreatment on the biogas potential of macroalgae biomass and the impact of co-digestion with other wastes (i.e. wheat, corn, grass, sugar beet vegetable mix, pig manure, and brewery spent grain/BSG) was explored. Paul et al. [71] conducted a series of Biochemical Methane Potential (BMPTs) Tests using a Biosystems AMPTS II system. The species *S. latissima*, collected from a macroalgae farm in Dingle Bay (Ireland) during the summer harvest in June 2016, was investigated for BMP in isolation and mixed with other waste materials at a ratio of 30:70. This study reported that *S. latissima* had a methane potential of 0.391 m^3 CH_4/kgVS and 0.472 m^3 CH_4/kgVS (mixed with wheat). Furthermore, Tabassum et al. [68] found that seasonal variation has impacted the composition (i.e. proximate, ultimate, and biochemical) of a typical Irish brown macroalgae species of *L. digitata*, influencing biogas production. Their studies further revealed that *L. digitata* harvested during August resulted in higher methane potential (0.327 m^3/kg VS) and higher biomass yields. These occurred due to the highest content of TS (19.7%5 ww), VS (81.7%TS), and C/N ratio (32). These findings confirmed that season and harvest time influenced the macroalgae's composition and characteristics, hence the biogas and methane yields.

4.3. Impact of chemical composition

Chemical composition variations in macroalgae are influenced by many environmental factors, including seasonal variability, light, water temperature, water salinity, and nutrient availability [27], hence influencing biogas and methane yields. For instance, Tedesco and Daniels [177] also reported the chemical compositions of five species of brown macroalgae (i.e. *Fucus serratus*, *Fucus vesiculosus*, *Ascophyllum nodosum*, *Laminaria digitata*, and *Laminaria saccharina*) were varied due to seasonal variability. The variations were evident in the organic matters, where an increase in %VS was correlated to an increase in methane yield. Montingelli et al. [27] found that *Laminaria sp* harvested in Autumn has much higher carbohydrates and VS content, resulting in higher methane yields than in the early Spring. Similarly, Adams et al. [62] found that *Laminaria digitata* harvested in July contain higher laminarin and mannitol content, producing more methane at 0.254 m^3/kg VS. The lowest methane yield was produced from *Laminaria digitata* harvested in March (0.196 m^3/kg VS), possibly due to the lowest carbohydrate content.

4.4. Impacts of co-digestion

Many studies have highlighted that co-digestion of macroalgae with other potential biomass substrates may affect methane and biogas yields. For instance, Paul et al. [71] demonstrated that biogas and methane yield production from cultivated macroalgae species *S. latissima* can be enhanced through co-digestion with various waste feedstocks. There was a synergistic effect on co-digestion of the biomass with all the biomass from different locations/seasons/growth types. Cultivated and wild *S. latissima* have yielded a high methane potential, counted at 0.393 m^3 CH_4/kgVS and 0.249 m^3 CH_4/kgVS, respectively. These studies further confirmed that *S. latissima* was suitable for biogas production in isolation and as a feasible novel feedstock to supplement co-digestion. The findings indicated that cultivated macroalgae have better methane production potential than wild macroalgae, highlighting the need to optimise cultivation methods further. Furthermore, FitzGerald et al. [141] investigated the AD process by treating *Ulva lactuca* co-digested with dairy wastewater at a ratio of 75:25 and 25:75, respectively. The AD was carried out under mesophilic conditions using CSTR for 40 weeks. The study revealed that *Ulva lactuca* could be converted into biogas with methane potential ranging from 0.174 to 0.177 m^3CH_4/kg VS. When a higher proportion of *Ulva lactuca* (at an OLR of ≥ 2 kg VS/m^3/day) was fed into the digester, this was led to digester instability due to the accumulation of VFA and overloading. Suhartini et al. [135] reported that anaerobic mono-digestion of wild *Gracilaria verrucosa* generates an SMP of 0.060 m^3 CH_4/kg VS. However, co-digestion of wild *Gracilaria verrucosa* with tofu dregs or food waste (at a ratio of 50:50) generated higher methane yields, with values of 0.112 and 0.165 m^3 CH_4/kg VS, respectively. Our previous study also reported that co-digestion of wild *Gracilaria verrucosa* with tofu dregs (at a ratio of 90:10 and 80:10 on a VS basis) increased methane production by 1.1–1.7-fold [18]. These findings confirm that macroalgae are potential feedstock in a single – or co-digestion system. However, in-depth investigations are still needed to optimise the process with a combination of sustainable pretreatment options.

In Scenario 2, it is widely known that glucose is critical as a carbon source in fermentation for producing bioethanol. Hence, the cellulose recovery or the production of sugars from carbohydrates hydrolysis is becoming the rate-limiting process [63]. The hydrolysis efficacy needs to be monitored and improved by adding pretreatments (i.e. acid or enzyme) to increase the process performance [178]. In bioethanol production, the challenges are often due to fermentation inhibitors, which need to be tackled, for example, by using activated carbon [101]. Scenario 2 proposed the integration of fermentation technology with the use of briquetting or pelletising technology. The technologies can convert the remaining solid residues from ethanol extraction, and they are considered simple and more economical options [179]. While in Scenario 3, the co-production of bioethanol with biogas, compost, and solid fuels seems to have more profitable alternatives. However, an increase in operational cost and the economic benefits from energy and bioproducts need to be considered when implementing this scenario. Other studies have highlighted that combining bioethanol and biogas production was the most feasible and suitable alternative due to a higher energy export potential than bioethanol or biogas alone [94]. Similarly, Ben Yahmed et al. [180] reported that converting green macroalgae *Chaetomorpha linum* with integrated biorefinery of bioethanol and biogas is feasible. Reusing the resulting waste or using locally produced enzymes for saccharification may reduce operational cost.

The above findings demonstrated that implementing a cascading and integrated biorefinery and closed-loop system is the most successful route for macroalgae conversion to bioenergy. Adopting scenario 3, which integrates fermentation technology (for bioethanol production as the primary process) with AD, composting, briquetting, and pelletising technology, could produce multi-products with high economic value. Macroalgae. However, further in-depth investigation is required, including optimising the cascading process, evaluating environmental impacts (by conducting a life cycle assessment/LCA study), and assessing the feasibility (by conducting techno-economic analysis/TEA).

5. Opportunities and challenges

Utilising marine macroalgae for bioenergy requires further in-depth studies to ascertain the cultivation process. Macroalgae cultivation may either depend on the natural stocks or on the artificial cultivation system to fulfil the macroalgae supply for energy and food purposes [32,163,181]. Various opportunities and challenges are evident in several aspects: supply chain, cultivation technology, potential market, and regulatory perspectives.

5.1. Supply chain

The macroalgae supply chain for bioenergy is critically important to ensure the feedstocks are renewable and sustainable. According to Ghadiryanfar et al. [6] and Sudhakar et al. [178], the supply chain of macroalgae for the production of bioenergy and high-value-added products, consisted of cultivation, harvest, transportation, pretreatment, conversion, and products, as shown in Figure 4. They explained that the cultivation technologies are commonly classified into naturally occurring macroalgae (wild macroalgae), aquaculture (cultured macroalgae), hatchery, and production either in vertical or horizontal systems. Macroalgae can be harvested mechanically with machinery or manually with human labour. Physical pretreatments are often preferred for macroalgae, including cleaning or removing foreign objects, drying/dewatering, particle size reduction (i.e. maceration, grinding, cutting), and desalination (i.e. washing with cold or hot water). The conversion routes can be classified as chemical (i.e. transesterification), biological (i.e. AD and fermentation), and thermochemical (i.e. pyrolysis, hydrothermal liquefaction, etc.). Finally, the products are composed of bioenergy (i.e. biogas, bioethanol, biodiesel, etc.) and high value-added products (i.e. biochemical, feeds, etc.). Macroalgae harvesting can be done by human labour (i.e. manual handling) and by machinery/harvesting equipment (i.e. mechanical) [6,178,182]. In Indonesia, our survey suggested that manual handling is widely adapted using a conventional equipment, such as small plastic boats or rafts to harvest macroalgae, as shown in Figure 5. Furthermore, drying is a critical step to reduce the water content, thus allowing longer storage time before selling to the macroalgae processing industry. In Indonesia, after harvesting, macroalgae are sun-dried (for 2–5 days) using 4 types of drying equipment (i.e. concrete slab with good drainage, tarpaulins or plastic on flat ground, flakes ('para-para') made of bamboo or plastic net, and wooden or bamboo rack for hanging macroalgae) [183]. An example of drying method for macroalgae in Indonesia, is shown in Figure 6. The drying process is commonly carried out near the cultivation pond or area to reduce energy consumption and operational cost.

, Large production is often required to enable a continuous supply of macroalgae and fulfil the industrial requirement. Thus, an appropriate storage system and cleaning and drying process are needed in the supply chain to maintain quality [182]. This challenge can easily be overcome since macroalgae harvesting was continuous over the year within a 2–5 month cycle [178]. However, Indonesia faces a challenge of natural disease (i.e. *ice-ice* disease) that leads to massive crop failure of *Eucheuma* genera and a decreased yield due to changes in salinity, sea surface temperature, and light intensity [183]. This disease could highly occur in Indonesia, with

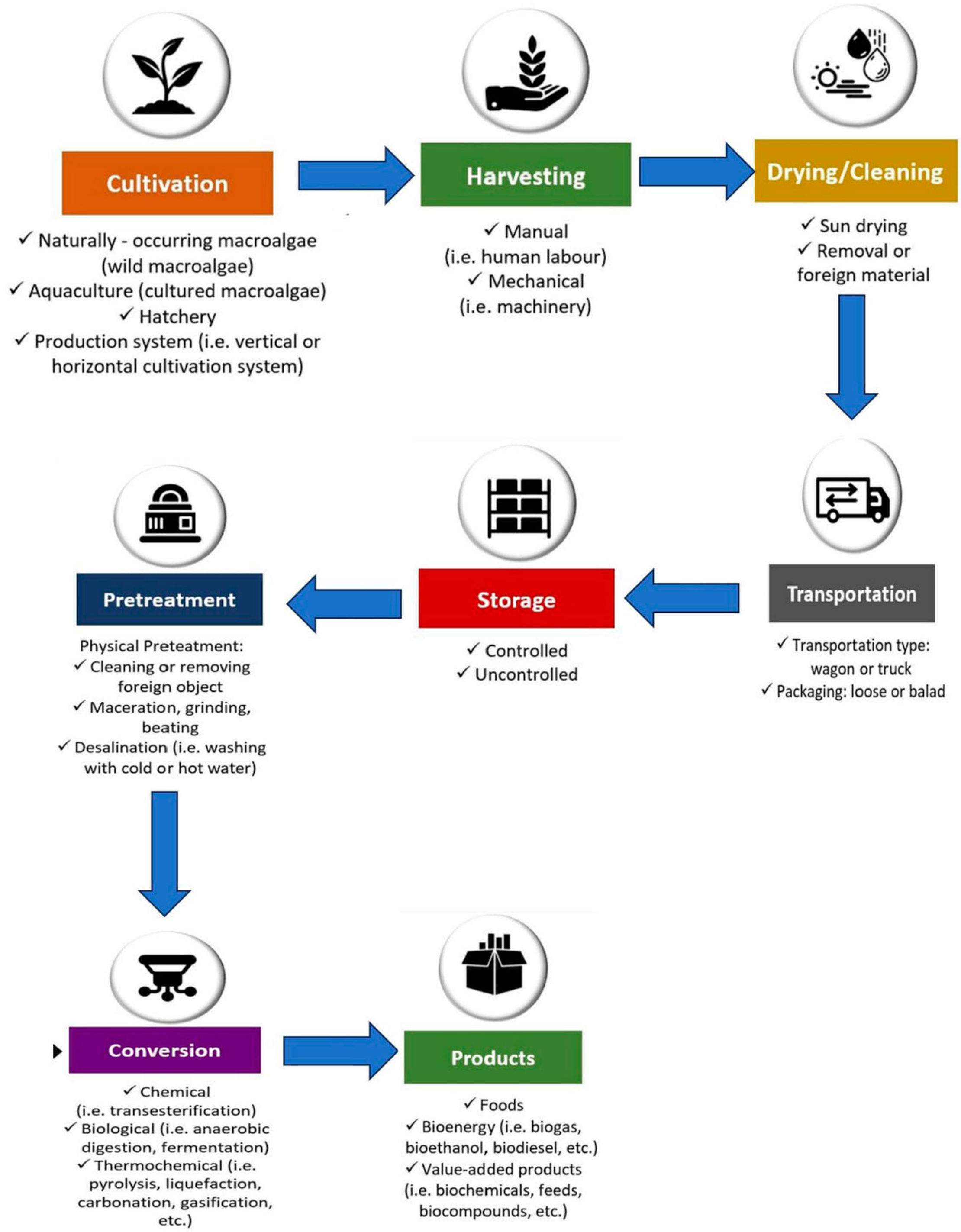

Figure 4. Supply chain of macroalgae for bioenergy and high-value added products.

estimation occurrence of 70–100%, thus leading to economic losses and influencing the price of macroalgae [184]. In addition, an appropriate drying/cleaning process and storage system are needed to maintain the quality, as well as the moisture content must be under 10% to avoid spoilage and improve shelf-life [185]. The post-harvest process is also a challenge that needs to be considered in the supply chain of macroalgae in Indonesia. According to Zamroni [186], West Nusa Tenggara Province still lacks investment in drying equipment and low awareness of quality control. Purnomo et al. [187] reported that, in Brebes regency as one of the primary locations for *Gracilaria* sp. cultivation, still have a lack of ability to control impurities due to poor post-harvesting treatment (especially during washing and drying).

Figure 5. Harvesting method for *Eucheuma cottonii* cultivation in Madura Island, East Java, Indonesia (Own photos).

Figure 6. Drying methods for *Eucheuma cottonii* (in Madura Island) and *Gracilaria verrucosa* (inSidoarjo City), East Java, Indonesia. (Own photos).

Then, in Serang Regency, the drying process was done by placing macroalgae on the ground, causing high impurities. According to Kumar et al. [188], various challenges and limitations during the cultivation, harvesting, and post-harvesting stages include seasonal issues, shortage of skilled labour for cultivation, low public acceptance of macroalgae farming, lack of automatic harvesting technology, high labour cost for harvesting, high cost for storage, low water quality, and lack of transportation.

5.2. Cultivation technology

Traditionally, macroalgae have been harvested from natural stocks or wild growth; however, these resources are depleted by overharvesting and non-scientific harvesting practices. Today, macroalgae cultivation has become more standardised, routine, and economical. Different factors such as morphology, regeneration capacity, and complex interactions (i.e. light/irradiance, temperature, nutrients, and water movement) are responsible for large-scale macroalgae cultivation. Different taxa require different farming methods; during the last five decades, almost 100 macroalgae taxa have been tested in field farms, but only a dozen are being commercially cultivated today [189]. Macroalgae cultivation is predominantly cultured by human activities (around 95%) and is commonly from the genera *Porphyra, Undaria, Laminaria*, and *Gracilaria* [190]. Furthermore, among the three varieties of red, brown, and green algae, brown algae (also known as kelp) is the fastest-growing class and contains 60% of carbohydrates (on a dry weight basis) [63].

Cultivating macroalgae consists of two stages: the hatchery and the grow-out phase [63,183]. The process is cyclical because the cultivation procedures start straight after harvest, and as the selection of fertile macroalgae is complete, the hatchery cultivation processes can be initiated. Macroalgae can be cultivated in different ways: offshore, onshore, or near-shore. Onshore cultivation is costly, and offshore cultivation has been known to be a better alternative. However, cultivating macroalgae requires high costs (i.e. capital, operational, maintenance, etc.) [63]. The study further claimed that AD of macroalgae is highly favoured as the process could use feedstocks with high moisture content. Commercial macroalgae cultivation has to consider physiological factors (i.e. salinity, temperature, nutrients, etc.) to produce a better biomass yield. Factors (i.e. transportation, operational cost, energy consumption, and process productivity) should be considered in macroalgae cultivation. According to van der Heijden et al. [183], in Indonesia, the capital investment for macroalgae cultivation typically ranges from US$ 311.40 – US $1,661, consisting of cost for inputs (i.e. first macroalgae culture, fertiliser, etc.), labour cost, energy cost, and equipment cost (i.e. boat, engine, drying apparatus, etc.). The transportation equipment for macroalgae cultivation including boats or rifts (made of bamboo or tyre). In contrast, vehicles transporting dried macroalgae to storage house include motorcycles, toa, or pick-up truck.

Numerous studies have explored the longer-term sustainability of the artificial cultivation of macroalgae. For example, near-shore sea or onshore waters macroalgae cultivation combined with the bioremediation principles of nitrates and phosphates removal may improve economic viability and sustainability [191]. In addition, methods such as sustainable long-line cultivation [192], integrated multi-trophic aquaculture (IMTA) where farming practices were combined with cultivation sustainably [193], indoors macroalgae photobioreactor (MPBR) [194], offshore macroalgae biorefinery systems [195], or a circulation and floating culture system (CFCS) [196], have also been investigated as a way of improving productivity, environmental sustainability, and its commercialisation. Rafael et al. [197] added that macroalgae cultivation could achieve more significant advantages by combining an integrated system involving sustainable farming practices and supporting policies to protect biodiversity (e.g. genetic conservation). Another study by Putri [198] has demonstrated the potential use of several macroalgae species (i.e. *Eucheuma spinosum*, *Padina minor*, and *Sargassum crassifolium*) for heavy metal removal, indicating its potential to combine macroalgae cultivation with bioremediation principles. Furthermore, macroalgae cultivation with IMTA application has been successfully implemented in Asia and Europe over the last two decades [199]. With the implementation of IMTA system, various benefits have been observed, including improved bioactive production and the growth of red macroalgae *Kappaphycus alvarezii* [200], increased yield of *Saccharina latissima* [201]; enhanced pharmaceuticals biofiltration to uptake enrofloxacin [202], improved the accumulation of oxytetracycline [203], enriched the identification of species suitability [204], and improved public image of sustainable macroalgae cultivation [199]. Macroalgae cultivation with IMTA system had become an alternative to minimise energy losses and reduce environmental deterioration. Thus, macroalgae cultivation can be more sustainable and profitable.

5.3. Policies and Regulations

Regarding regulatory practices and applications, Developed countries, such as the European Union, have better legislation that covers the macroalgae end use (i.e. medicine, foods, feeds, cosmetic products, packaging materials, fertiliser/biostimulant, and biofuels), aquaculture subsidies, aquaculture licensing, maritime spatial planning, tax schemes, etc. [205]. These rules consider all stakeholders involved in the macroalgae business; for example, legislation on products end use also has implications on the producers (i.e. cultivation and harvesting of macroalgae). These were found to impact the macroalgae business and its sustainability strongly. Macroalgae cultivation provides economic, social, and environmental benefits, including additional income, job creation, carbon sequestration, greenhouse gas (GHG) reduction, and nitrogen amelioration [195]. However, there are many challenges in creating sustainable macroalgae acquisition and valorisation to bioenergy; hence, legislation and policies to cover the matter are essential. For example, in the UK, two permissions (i.e. a lease from The Crown Estate and a marine license from the relevant regulator) must be obtained prior to any development activities to the marine environment. Such practices ensure that potential environmental impacts or risks to the local environment can be avoided or mitigated [206]. Wood et al. [206] suggested that a detailed review of the cultivation's environmental and social aspects, as well as the legislative and regulatory frameworks which support or inhibit current practice is critical. This review is required to avoid or eliminate the major negative environmental impacts of the practice. They argued that the current legislation or guidance needs to be updated to cover the cultivation and valorisation of macroalgae. They recommended that a lease and a Marine license are compulsory for the relevant producers or stakeholders.

Tthe Indonesian Government has set of regulations and standards procedures to support the sustainability of the macroalgae business, including aquaculture farm management regulations (Presidential Regulation No. 28 Year 2017), macroalgae seed quality checking (SNI No. 7672/2011), and macroalgae cultivation method (SNI No. 7579/2010) [207]. The Indonesian Government has also provided guidelines on the National Roadmap for developing macroalgae industries (regulated in Presidential Regulation No. 33 Year 2019, emphasising the cultivation to the macroalgae processing stages) [208]. However, no regulations and policies covered macroalgae valorisation for bioenergy or biochemical products. The regulation on biofuel development is stated in Law No. 30 Year 2007 on Energy, which underlines energy sources' diversification, conservation, and intensification [209]. Other regulations have highlighted the development of new and renewable energy (NRE) but still lack mention of marine biomass. These regulations include Government Regulation No. 79 Year 2014 on National Energy Policy (NEP), Presidential Regulation No. 22 Year 2017 on General Planning for National Energy (GPNE), and MEMR Regulation No. 12 Year 2015 on Biofuel Supply [210,211]. India, as a developing country, has also faced problems with legal and regulatory aspects on biorefining macroalgae for foods, biochemicals, and biofuels, especially in support of the Government to accelerate research and development in cultivation, harvesting, processing, and commercialisation of macroalgae [212]. Other challenges include regulating and licensing aquaculture and biofuel applications [188].

5.4. Market potential

Many studies have reported alternative market opportunities for macroalgae to produce bio-based chemical compounds. For example, Irianto and Dewi [213] reported that Indonesia is one of the largest countries producing macroalgae with economic benefits, including *Gracilaria* and *Gelidium* (for agar production), *Laminaria* and *Sargassum* (for algin production), *Hypnea, Eucheuma,* and *Kappaphycus* (for carrageenan production). However, this study reported many other species, such as *Turbinaria sp.*, *Ulva sp.*, and *Laurencia sp.*, which have high polyphenol and carotenoids beneficial as free radical-scavenging properties. Further research on the potential for anticancer nutraceuticals was highlighted. Widowati et al. [214] stated that three Indonesian native macroalgae (i.e. *Sargassum echinocarpum*, *Sargassum duplicatum*, and *Sargassum polycystum*) have a great potential to be valorised into antibacterial and radical-scavenging compounds due to their high alkaloid and flavonoid content. Furthermore, Nursid et al. [185] identified 20 macroalgae species locally from Binuangeun Beach, Kabupaten Lebak, Banten, Indonesia. The species include 7 species of Chlorophyta (i.e. *Chaetomorpha sp., Caulerpa sertularioides, Chaetomorpha antennina, Ulva reticulata, Ulva fasciata, Halimeda macroloba,* and *Caulerpa racemosa*), 9 species of Phaeophyta (i.e. *Padina australis, Sargassum echinocarpum, Turbinaria ornata, Turbinaria conoides, Sargassum ilifolium, Turbinaria decurrencs, Sargassum polycystum, Sargassum binderi, Homophysa triquetra*), and 4 species of Rhodophyta (i.e. *Hypnea* sp., *Gracillaria edulis, Gigartina chauvanii,* and *Spatoglossum solieri*). The study reported that all species have a high antioxidant activity determined from their polyphenol content. The highest polyphenol was extracted from *Padina australis* (58.59 mg GAE/g), followed by *Caulerpa sertularioides* (28.31 mg GAE/g) and *Hypnea* sp. (22.05 mg GAE/g), respectively. Takarina and Patria [215] studied the potential of polyphenol or phenolic compounds from Indonesian macroalgae (e.g. *Caulerpa racemosa*) obtained from Pasir Gosong Lebar Beach, Indonesia. The study found that *C. racemosa* contains 2.57–4.58% polyphenol compounds, mainly catechin and tannin. These compounds are famous for their medical benefits. This study further suggested that *C. racemosa* can be used to produce antibacterial, antiseptic, and antioxidants.

Furthermore, macroalgae can be converted into organic acids, such as volatile fatty acids (VFA) (i.e. acetic acid, propionic acid, succinic acid, butyric acid, etc.). Pham et al. [216] examined AD technology for producing VFA from three marine macroalgae (i.e. *Laminaria japonica*, *Pachymeniopsis elliptica*, and *Enteromorpha crinite*). The results demonstrated that acetic, propionic, and butyric acids were the main VFA components produced in the highest quantities. *Laminaria japonica* produced VFA at the maximum value of 15.2 g/l from 50 g/l macroalgae sample when incubated at 35°C and pH 6.5–7.0 for three days. These findings confirmed that macroalgae might be a potential feedstock for VFA production. Other studies have also shown the potential production of acetic acid and succinic acid from marine macroalgae, such as *Gelidum amansii* [217], *L. digitata* [54], *S. latissima* [40], *Sargassum muticum* [218], *Undaria pinnatifida* [219], *Ulva* spp. [220], *Enteromorpha intestinalis* [221], and *Gracilaria vermiculophylla* [222]. Combination with pretreatment was found to improve organic acid production from macroalgae. Pham et al. [216] reported that VFA production from *L. japonica* increased by 56% with the alkaline and thermal pretreatment. While Pham et al. [223] found biological pretreatment of *L. japonica* using *Vibrio* sp. resulted in an increase in VFA production by 88% (from 8.3 g/L (untreated) to 15.6 g/L (treated)). Another alternative pretreatment was using engineered *E. coli* to increase the succinic acid production from *L. japonica* [224] or *Palmaria palmata* [225]. Xia et al. [226] claimed that co-fermentation of microalgae (e.g. *Arthrospira platensis*) with macroalgae (e.g. *L.* digitata) improved VFA production by 33.2%. Ra et al. [227] found that hyper-thermal acid hydrolysis and adsorption pretreatment allowed efficient butyric acid production from *Gelidium amansi*. Several studies reported using macroalgae for high-value chemicals (i.e. VFA) in markets. For example, Obata et al. [228] investigated the hydrolytic pretreatment impact on the VFA from *L. digitata* and *Ascophyllum nodosum* species. Two different pretreatments were explored, including acid only and acid followed by enzymatic pretreatment prior to the AD process. The results suggested that acid pretreatment with and without enzyme addition improved the effectiveness of the hydrolysis process, producing more VFA than untreated samples. These findings indicated that the AD of macroalgae could also be applied for producing VFA instead of biogas, with the operational parameter adjustment. A previous study by Milledge et al. [218] stated that *Sargassum muticum* is considered to have no economic value due to its heavy metal content. However, they argued that, by using aquaculture cultivation technology, *S. muticum* can be a potential source of health products considering its higher antioxidant level (i.e. tocopherol, fucoxanthin, other carotenoids or phenolic compounds) and organic acids.

While in Indonesia, the research and development focus on the valorisation of macroalgae for organic acid production is still lagging. The information and the growth of a bioeconomy based on diverse and prevalent macroalgae are still limited in the literature. Therefore, further work on Indonesian markets is required regarding the market potential for higher value streams from macroalgae.

6. Future prospects

Indonesia currently has a great potential for marine and cultivated macroalgae. Macroalgae farming in Indonesia, established in the 1990s, is continuously expanding and bringing the country as one of the largest macroalgae (or seaweed) producers. Various types of macroalgae are well cultivated in Indonesia for various purposes, for example, *Gracilaria sp.* and *Gelidium sp.* (agar production), *Laminaria sp.* and *Sargassum sp.* (algin production), or *Hypnea sp.*, *Kappaphycus sp.*, and *Eucheuma sp.* (carrageenan production). Many studies have also highlighted a vast prospect of macroalgae for food, cosmetic, and biomedical applications due to their high content of bioactive compounds [213]. As previously explained, macroalgae is a viable, feasible, and renewable resource for bioenergy production. Therefore, macroalgae have provided economic benefits to the country. Although many macroalgae are available in Indonesia, this biomass must be explored and utilised for bioenergy.

In Indonesia, the future application of macroalgae for bioenergy production should consider the sustainable integrated biorefinery concept for circular economy achievement. Kumar et al. [188] stated that the macroalgal sustainable biorefinery should address various aspects, including the scalability of cultivation, socio-economic benefits and impacts, reduction of adverse environmental impacts (through sustainable cultivation process and waste biorefinery), the green pretreatment options, the conversion technology, the geographical distribution of macroalgae, the collaboration of relevant stakeholders, and circular economy concept. Regarding macroalgae for bioenergy, the conversion technology should be selected based on the efficiency, efficacy, feasibility, and environmentally sustainable perspective. Compared to other conversion technologies reviewed here, anaerobic digestion (for biogas production) and fermentation (for bioethanol production) offer better potential. Combined with integrated biorefinery and circular economies, these conversion technologies can provide economic, social, and environmental benefits to local communities and relevant stakeholders. A previous study by Rajak et al. [229] highlighted that the cascading biorefinery approach of edible and non-edible macroalgae for multiple product generations increases the efficiency of resource valorisation and the elimination of residual waste. Their study emphasised the benefits of using residual macroalgae from bioethanol fermentation for other bioenergy resources (i.e. biogas, bio-oil, syngas, and biochar) and bio-products (i.e. biofertilizer). A study by González-Gloria et al. [230] stated that the macroalgal biorefinery, which integrates the cultivation system with bioethanol and biogas production, can provide additional economic benefits from bioenergy and biofertilizer production. Hence, integrated biorefinery and circular economy approaches can contribute to the sustainable utilisation of macroalgae, zero waste generation, and better socio-economic benefits.

7. Conclusion

Although the expertise regarding cultivation methods is progressing, Indonesia still faces problems, such as lack of knowledge and experience in efficient and cost-effective downstream processing. The generation of energy and high-value-added products from macroalgae could contribute to economic development in Indonesia and provide new business opportunities within a growing global bioeconomy. Understanding the sociocultural impacts and the wider environmental impacts of accelerating the development of this market is needed to ensure the longevity and sustainability of this sector. This review confirmed that macroalgae in Indonesia have the potential as feedstock for bioenergy production due to their availability (i.e. simplicity and sustainability of cultivation) and biochemical characteristics. Considering the technological readiness level and operational cost, implementing anaerobic digestion or bioethanol fermentation is a potential alternative to other bioenergy conversion routes in Indonesia. This review has demonstrated that the prospective and sustainable route to valorise macroalgae for bioenergy production is Scenario 3. This proposed scenario allows the integration of fermentation technology with other potential technologies, such as anaerobic digestion composting, briquetting, or pelletising solid residues. Hence, multiple products can be produced, including bioethanol, biogas, compost, briquettes, or biopellets. Thus, biorefining macroalgae to bioenergy could achieve zero-waste and closed-loop systems, where the process is operated sustainably with minimal or no waste generation.

Author contribution

Sri Suhartini: Conceptualization, Writing – original draft, Writing – review & editing, Visualisation, Supervision. **Martasari B P**: Writing – original draft, Literatures curation, Writing – review. **Elviliana**: Writing – original draft, Literatures curation, Writing – review. **Novita A R**: Writing – original draft, Visualisation, Writing – review. **Mochammad A J**: Conceptualization, Writing – review. **Roshni P**: Conceptualisation, Writing – original draft. **Irnia N**: Conceptualization, Writing – review, Supervision. **Nur L R:** Conceptualization, Writing – review, Visualisation. **Lynsey M**: Conceptualisation, Writing – review & editing, Supervision.

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