



# Optimisation of biogas generation from brown seaweed residues: Compositional and geographical parameters affecting the viability of a biorefinery concept

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## HIGHLIGHTS

- Biorefined seaweed residues demonstrated high potential for producing biogas.
- Ambient extractions cascade improved the CH<sub>4</sub> and biodegradation potentials.
- Average CH<sub>4</sub> yields on an annual basis were found between 107 and 405 mL gVS<sup>-1</sup>.
- Seasonal composition and harvest site greatly affect digestion performance.

## ARTICLE INFO

### Keywords:

Macroalgae residues  
Extraction  
Integrated biorefinery  
Methane potential  
Acclimatation  
Anaerobic biodegradability

## ABSTRACT

Very recently, integrated biorefinery approaches are being developed with the aim to produce high-value products for a variety of industries in conjunction with green energy from sustainable biomass. Macroalgae (seaweed) have been regarded as more sustainable compared to terrestrial crops, since they do not occupy land for growth. Macroalgal biomass changes greatly according to species and harvest season, which affects its chemical energy potential. This study was conducted seasonally on five species of brown seaweed over a yearlong period to investigate the effects of chemical composition variations, bioproducts extraction processes and inoculum acclimatation on methane production. As a result of the bioproducts extraction, it was found the seaweed residues exhibit a great potential to produce methane. Stoichiometric methane yield and C:N ratio changed in favour of an improved digestibility with bioconversion rates greater than 70% in some instances, i.e. achieved by *Laminaria* species and on the West coast *Fucus serratus*. The two *Laminaria* species investigated also presented the highest CH<sub>4</sub> production rate, with *Laminaria digitata* reaching 523 mL CH<sub>4</sub> gVS<sup>-1</sup> and *L. saccharina* peaking at 535 mL CH<sub>4</sub> gVS<sup>-1</sup> with acclimatised and non-acclimatised sludge respectively.

## 1. Introduction

Seaweed biomass has been under the spotlight as feedstock for biogas production in the recent years. Seaweeds (or macroalgae) are regarded as third generation feedstocks for biofuels, since their use as energy crops exhibits several advantages when compared to terrestrial crops [1]. In particular, they are not quite used as food source on a global scale, which minimises the impact on price related to the food versus fuel debate for first generation feedstocks, e.g. corn or palm oil. Furthermore, unlike second generation lignocellulosic crops such as wood, maize or grass, cultivation of macroalgae does not occupy arable

land. This translates into multiple benefits for cultivating marine crops. These are low or absent in lignin content (recalcitrant to biofuels conversion), no fresh water or nutrients provision is needed for growth. Also, faster growing rates than land crops and higher CO<sub>2</sub> remediation potential have been reported due to a more efficient photosynthesis [2]. Nonetheless, seaweed conversion to biofuels encounters several technical challenges such as seasonal variation in composition also depending upon geographical location [3] and necessity to undertake assessments of the impact of systematic wild-harvesting on marine ecosystems. Hence, cultivation techniques need to be improved or re-designed to fulfil specific species' requirements locally.

Acronyms: AD, Anaerobic Digestion; BI, Biodegradability Index; COD, Chemical Oxygen Demand; TS, Total Solids; VS, Volatile Solids

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<https://doi.org/10.1016/j.apenergy.2018.06.120>

Received 22 February 2018; Received in revised form 21 June 2018; Accepted 22 June 2018

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The EU has underpinned that seaweed biogas or biomethane as transport fuel may be playing a significant role for energy generation in the near future [4]. However, the main obstacle to harvesting the seaweed-to-energy potential lies in the vast volumes of biomass required to generate meaningful energy contribution to help the shift towards replacement of fossil fuels. A very interesting study by Allen et al. [5] on seaweed gaseous biofuels suggests its feasibility would be possible if suitable volumes of feedstock are obtained via aquaculture. It is yet unknown how this can be achieved sustainably as aquaculture techniques developed so far are not cost-effective to justify the use of this resource solely for energy purposes. A review by Ghadiryanfar et al. [6], identifies that, despite being more cost-competitive than other renewables, biofuels and bioenergy from macroalgae entail higher costs than terrestrial biomass due to costly cultivation. In their study, in fact, Roesijadi et al. [7] regards this as a key issue. The authors also identify that biogas production from macroalgae is more technically-viable than for other fuels however, the cost of marine crops needs to be reduced by 75% of the present level to make macroalgal biogas economically-feasible. The feasibility of algal biofuels can be significantly enhanced by a high-value co-product strategy [8] using an integrated biorefinery approach to produce simultaneously bioproducts and biofuels to enable circular economies. A comprehensive review by Jung et al. [9] on potentials of macroalgae as feedstock for biorefinery, reports that macroalgal biomass is currently utilized to source human food, algal hydrocolloids, therapeutic materials, fertilizer, and animal feed. The food industry, whose market share is 83–90% of the total seaweed industry, is the largest and accounts for \$5 billion worldwide on an annual basis [7]. This means that the remaining 10–17% used for extraction of bioproducts would be available to explore integrated biorefinery opportunities. In fact, when processed for extraction of bioproducts, a significant amount of sugar-rich seaweed residue is generated, which can easily be used for feeding anaerobic digesters. A study by Tedesco and Stokes [10] has investigated the biogas potential from macroalgal biorefined residues in October harvested in Co. Clare, Ireland. The authors identified a biogas potential between 182 and 453 mL gVS<sup>-1</sup>, with best results achieved from *Laminaria* spp. although they have not analysed the effect of seasonal variation on such yields.

Seasonal variation in composition has a major influence in determining the methane potential from marine biomass. Therefore, in order to obtain a stable biogas production, investigations are needed over a year period to assess the variability of the methane yields achievable and plan for complementary co-substrates for digestion. Very few studies have been conducted on seasonal composition of macroalgae [11–13]. These, however, have not investigated biorefined algal residues but have rather characterised the biomethane potential from freshly harvested or drift seaweed biomass. Nizami et al. [14] reported that the selection or integration of biorefinery technologies should be based on its waste characterisation. As biochemical characterisation changes seasonally in the fresh feedstock, it is expected that algal residues will also present a changing composition depending upon harvesting periods. Seasonal variation in composition was also found to represent one of the major technical challenges for seaweeds in the biobased economy by van Hal et al. [15].

Brown macroalgae have been selected for this research as these are mainly used in Irish industrial applications across a variety of sectors. Ireland's seaweed-based industry consists of small and medium businesses involved in production of animal nutrition, animal hygiene, plant health, soil fertilizers, alginate, cosmetics and nutraceutical products [16]. The Irish Fishery Board (BIM), reported that the Irish seaweed production and processing industry will be worth €30 million per annum by 2020 [17]. The waste products generated by this growing industry are not currently characterised for biofuels production. The literature heavily lacks of investigations examining the seasonal biogas potential from the algal waste streams derived from the existing bio-industry. Since feasibility studies on biogas generation from waste solids and liquids from seaweed processing plants are also relevant to

government authorities [18], this study aimed at characterising the methane yield response from the most common Irish brown seaweed residues generated by the local bioindustry.

Anaerobic digestion (AD) essays have been conducted over a year-long period during which brown seaweed biomass has been wild-harvested seasonally at two opposite sides of the island, in order to evaluate the influence of geographical location on composition. These are Howth Bay, Co. Dublin on the East coast and a number of bathing beaches in Co. Galway and Co. Clare on the West coast.

The harvested feedstocks underwent bioproducts extraction using room temperature extraction procedures provided by the project industry partner (Irish Seaweed Processors Ltd.) based in Ireland. The extraction processes used in this research also follow the seaweed biorefinery concept proposed by Balina et al. [19] in which polysaccharides, antioxidants, pigments and proteins are targeted by the extraction cascade that precedes a biogas production step, which utilises the leftover residue as input feed for AD. The biochemical composition (1) was analysed after collection of the biomass and again following bioproducts extraction to identify the residual organics content. Effective methane yields (2) and biodegradability indices (3) against the theoretical stoichiometric yields were used to evaluate the methanogenic potential against the actual methane yields from the feedstocks. As the resulting pH of suspended residues solution was highly alkaline, acclimatation (4) of the inoculum for improved gas yields was also tested and yields compared with performance of non-acclimated inoculum.

## 2. Materials and methods

### 2.1. Substrate collection and inocula

Biomass of *Fucus serratus* (FS), *Fucus vesiculosus* (FV), *Ascophyllum nodosum* (AN), *Laminaria digitata* (LD), and *Laminaria saccharina* (LS) was collected seasonally at low tide (2015–16) and underwent extraction at room temperature of bio-compounds at laboratory scale, as per procedure provided by the industry partner (Irish Seaweed Processors Ltd). The extracting procedure adopted by the processing company targets the extraction of alginic acid, fucoidan, fucosantoin, laminarin, mannitol, and proteins.

The collections took place on the East and West coasts of Ireland in order to investigate the effect of geographical location on the biomass composition. The harvesting sites were Co. Galway and Co. Clare beaches on the West side of the island with collections in May, September, November and January, and Howth Bay on the East side with collections in June, October, November and January. Samples were harvested and frozen within 24 h to  $-20\text{ }^{\circ}\text{C}$  until use. The collections started in May/June 2015 and were completed in January of the following year.

In order to add the necessary fermenting microorganisms to the reactors, the residue samples were then incubated with 300 g of digested sewage sludge, provided by the wastewater treatment plant of Celtic Anlian Water (CAW) Ltd. The initial sludge's pH was measured as  $8.1 \pm 0.02$ . Acclimatation was conducted by inoculating reactors with extracted residue of the same seaweed species to be subsequently digested and allowing fermentation to occur for approximately 10 days before incubation in the reactors.

### 2.2. Proximate and ultimate analysis

Dry organic matter or Total Solids (TS) and Volatile Solids (VS) contents were determined by using a high-temperature oven via overnight drying of the samples at  $105\text{ }^{\circ}\text{C}$ , followed by combustion at  $575\text{ }^{\circ}\text{C}$  of the seaweed residues, as by standard procedure [20]. Tests were conducted in duplicate.

The ultimate analysis was outsourced to Celignis Ltd. (Irish biomass laboratory) to identify the elemental composition of the fresh and residual substrates. The carbon, hydrogen, nitrogen, and sulphur contents

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