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Waste paper and macroalgae co-digestion effect on methane production

Cristina Rodriguez [a,*], Abed Alaswad [b], Zaki El-Hassan [a], Abdul G. Olabi [a]

[a] Institute of Engineering and Energy Technologies, School of Engineering and Computing, University of the West of Scotland, Paisley, PA1 2BE, UK
[b] School of Engineering and the Built Environment, Birmingham City University, Birmingham, B5 5JU, UK

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ABSTRACT

The present study investigates the effect on methane production from waste paper when co-digested with macroalgal biomass. Both feedstocks were previously mechanically pretreated to reduce their particle size. The study was planned according two factors: the feedstock to inoculum (F/I) ratio and the waste paper to macroalgae (WP/MA) ratio. The F/I ratios checked were 0.2, 0.3 and 0.4 and the WP/MA ratios were 0:100, 25:75, 50:50, 75:25 and 100:0. The highest methane yield (386 L kg^{-1} VS$_{added}$) was achieved at an F/I ratio of 0.2 and a WP/MA ratio of 50:50. A biodegradability index of 0.87 obtained in this study indicates complete conversion of feedstock at an optimum C/N ratio of 26. Synergistic effect was found for WP/MA 25:75, 50:50 and 75:25 mixing ratios compared with the substrates mono-digestion.

1. Introduction

EU and UK Government have tightened their waste disposal regulations, landfill disposal of organic waste will be no longer available after 2020 [1], so alternatives to waste disposal on landfills are required for an efficient and profitable recycling. By the same year of 2020, EU aims to get the 20% of energy consumption from renewable resources, 10% coming specifically from biofuels [2,3].

Waste management and energy recovery can be effectively combined in the anaerobic digestion process. Anaerobic digestion performed under controlled conditions also allows pollution reduction and emissions control, reducing greenhouse gas emissions compared to fossil fuels by the utilization of local resources [4]. Biogas is obtained from waste materials through the anaerobic digestion process. In the same process, a by-product with fertilizer value is obtained (the digestate) [5–7]. Upgraded biogas, named biomethane, with a concentration greater than 97% can substitute natural gas in Combined Heat and Power Plants (CHPP) and may be injected into the gas grid or compressed and used as transport fuel [8].

Paper and cardboard account for 25–30% of municipal solids waste (MSW) [9,10]; the biggest source of waste paper is industry and businesses with the 52% of the total [11]. Anaerobic digestion of waste paper is usually studied as part of the anaerobic digestion of MSW. In some cases, the study was carried out on the MSW different fractions that resulted in methane yields for newsprint paper from 58 to 100 L kg^{-1} VS$_{added}$ [9,12]; for office paper 208–369 L kg^{-1} VS$_{added}$ [9,12–15] and for cardboard 96 and 217 L kg^{-1} VS$_{added}$ [9,15].

The ratio carbon/nitrogen (C/N) is one of the most important factor in anaerobic digestion nutrients balance. Carbon is the source of energy for the process and nitrogen is needed for the formation of enzymes that perform metabolism. A high C/N ratio is an indication of rapid consumption of nitrogen by methanogens and results in lower gas production, while a low C/N ratio causes ammonia accumulation and pH rises excessively. Most authors consider an optimal C/N ratio needs to be in the range 10–30 [4,16,17]. Considering other macronutrients, the C:N:P:S ratio in the reactor should be 600:15:5:3 [16]. Paper materials have a carbon-to-nitrogen (C/N) ratio ranging from 173/1 to greater than 1000/1 [18], these values are very high for anaerobic digestion so a balance of nutrients can be achieved through co-digestion with biomass that contains nitrogen and lower the C/N ratio. Digestion of nitrogenous substrates (C/N ratio less than 15) can lead to

Abbreviations: AD, anaerobic digestion; ANOVA, analysis of variance; F/I, feedstock/inoculum; KDP, potassium dihydrogen phosphate; MC, moisture content; RSM, response surface methodology; TS, total solids; VS, volatile solids; WP/MA, waste paper/macroalgae.

* Corresponding author.
E-mail address: cristina.rodriguez@uws.ac.uk (C. Rodriguez).

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problematic digestion caused by excess levels of ammonia, increasing the pH levels in the digester leading to a toxic effect on methanogens population [19,20].

Co-digestion is the simultaneous digestion of a mixture of two or more substrates and offers many advantages, including ecological, technological, and economic benefits, compared to digestion of a single substrate [21]. The purpose of co-digestion is usually to balance nutrients (C/N ratio and macro- and micronutrients) and dilute inhibitors/toxic compounds. Moreover, the co-digestion of two or more complementary substrates may induce a synergetic effect on their biodegradability, causing an increase in the methane yield and production rate [22]. Zhong et al. achieved maximum methane yield in co-digestion of algae and corn straw at C/N ratios between 20 and 25 [23]. Co-digestion of waste paper with *Scenedesmus spp*. and *Chlorella spp*. achieved a maximum methane yield at a C/N ratio of 18 [24].

Further advantages of co-digestion include the unification of feedstock's management by sharing treatment facilities, reducing investment and operating costs. Successful examples of co-digestion include: cow dung and water hyacinth [25]; algal sludge and waste paper [26]; cattle manure and crude glycerine [27]; grass and sludge and [28]; municipal sludge, microalgae and waste paper [4]; algae biomass residue and lipid waste [29] and hay and soybean [21].

Co-digestion can result in a positive effect (synergistic effect) on the degradation of each individual substrate in the mixture and/or an increase in the methane yield kinetics [30]. This improvement may arise from the contribution of additional alkalinity, nutrients, enzymes and trace elements that a feedstock by itself may lack and an increased buffering capacity. Evenly allocated nutrients in co-digestion would support microbial growth for efficient digestion, while increased buffering capacity would help maintain the stability of the anaerobic digestion system [31]. Antagonistic effects may result from low C/N ratios resulting in high total ammonia nitrogen (TAN) released and high volatile fatty acids (VFAs) accumulated in the digester leading to a suppression in the cellulase activity and a decrease in the methane yields. Antagonistic effects can come also from other several factors, such as pH inhibition and ammonia toxicity [32]. Synergistic effects were found on the co-digestion of primary sludge and paper pulp reject with an improvement of 32% on methane yield [33] and the co-digestion of Taihu blue algae with corn straw (up to 60% extra methane) [31].

The innovation in this study is that it is the first to assess the optimized conversion of waste paper to biogas through co-digestion with macroalgae (*P. canaliculata*) as a source of nitrogen to balance the C/N ratio in the process. Macroalgae is a great source of biomass in Scotland and its optimization as a feedstock for anaerobic digestion is being addressed. The optimization include both pretreatment and co-digestion for a final improved methane potential. Both feedstock were previously mechanically pretreated in a Hollander beater according to [34,35]. The study was planned to check different levels of feedstock/inoculum ratio (F/I) and waste paper/macroalgal (WP/MA) mixing percentages. A statistical analysis through Response Surface Methodology (RSM) is presented to provide a more comprehensive evaluation of the interaction between the process parameters on the methane production.

2. Materials and methods

2.1. Feedstock and inoculum

Pelvetia canaliculata, a brown macroalgae commonly known as channelled wrack, was collected on-shore (55°55′ N 5°09 W) in the Isle of Bute, Scotland in March 2016, refrigerated at 4 °C and used within 4 days. Mature specimens were chosen of minimum length size of tufts of 10 cm. Small contaminants like plastic or stones were removed but the algae was not washed as the algae is considered in this study a waste material to be used as found in the shore. Waste paper was collected from recycle bins at the School of Computing and Engineering at the University of West of Scotland (UWS) in Paisley, Scotland. Feedstock characterization was shown in Table 1. Both feedstocks were previously mechanically pretreated in a Hollander Beater, the optimized time of pretreatment for macroalgae was 50 min and for waste paper was 55 min. During the pretreatment, the biomass is mixed with water and a pulp is produced, this pulp is directly fed the reactor to help to fluidizer the process. Table 1 details the characterization of the macroalgae and the waste paper.

The sludge used as inoculum was provided by the Strathendrick Biogas Plant (Balfron, Scotland) which used dairy farm cow slurry, distiller's draff and pot ale syrup from local whisky distilleries and some grass silage as feedstock. The inoculum was refrigerated at 4 °C and used next day of collection (total solids (TS): 7.59%, volatile solids (VS): 88.63%, ash content: 11.37%). Total and volatile solids (TS, VS) of both feedstocks and sludge were calculated in duplicate and were obtained submitting random samples of pretreated biomass at 105 °C (for TS) and 550 °C (for VS) until constant weight. The VS are expressed as percentage of TS.

2.2. Biomethane potential test

The biomethane potential test were set according [36,37]. Erlenmeyer flasks of 0.5 L with a working volume of 0.4 L were used as bioreactors; the biogas was collected in airtight Linde PLASTIGAS bags. Nitrogen was flushed into the headspace of each reactor to preserve the anaerobic conditions and clear up any trace of oxygen from the system. The bioreactors were placed in a water-bath to maintain the mesophilic temperature at 37 °C.

Reactors were fed with a fixed amount of 200 g of sludge (inoculum) and the quantities of macroalgae and waste paper pulp required to meet the feedstock/inoculum (F/I) ratios (0.2, 0.3 and 0.4) and the waste paper/macroalgae (WP/MA) ratios (0:100, 25:75, 50:50, 75:50 and 100:0). The F/I and WP/MA ratios are represent in terms of VS. Control batches were prepared in the same way except for the feedstock addition to assess the inoculum contribution of the methane production. The pH was adjusted to 6.95 ± 0.40 with potassium dihydrogen phosphate (KDP) as a buffer solution. To facilitate the contact biomass-inoculum and degasification of the substrate, flasks were daily shaken during the process. The gas volume was measured with an upside-down cylinder connected to a bubbling flask to maintain anaerobic conditions; the methane content was test with a gas analyser (Drager X-Am 7000). Average results were reported in this paper from duplicated tests in terms of mL of methane per g of VS added of feedstock. Methane yields are given for a dry gas in standard conditions of temperature (0 °C) and pressure (1 atm).

Table 1
Feedstock characterization.

Parameters	Macroalgae	Waste paper
Total Solids (%)	6.17 ± 0.13	2.55 ± 0.02
Volatile Solids (% of TS)	80.18 ± 0.05	97.30 ± 0.07
Ash content (%)	19.82 ± 0.05	2.70 ± 0.03
Carbon (% of TS)	38.15 ±	36.87 ±
Hydrogen (% of TS)	5.48 ±	3.61 ±
Nitrogen (% of TS)	2.63 ±	0.30 ±
Oxygen (% of TS)	34.32 ±	56.52 ±