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Biological pretreatments of biomass for improving biogas production: an overview from lab scale to full-scale



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ABSTRACT

Recent shifts in European countries biogas policies tend to limit the use of energy crops and encourage the use of manure, lignocellulosic feedstocks and bio-waste. The need to use feedstocks that are more difficult to handle (displaying either too low or too high biodegradation rates) is calling for the development of adapted pretreatments. Among them, biological pretreatments are very promising due to their reasonable cost, environmental friendliness and possible application to a wide spectrum of feedstocks. They can be divided into three categories: enzymatic, anaerobic and aerobic ones. This review aims at providing some guidelines on which type of biological pretreatment to apply for a given feedstock. To deliver such recommendations we considered the full range of technological readiness level. We gathered an analysis of the recent literature data obtained at lab or pilot scale focusing on methane yield enhancements and the description of some full-scale commercialized technologies. For lignocellulosic feedstocks, both enzymatic pretreatments using lignin-modifying enzymes or carbohydrases and aerobic pretreatments using consortia or simple aeration appear as promising. For bio-waste, anaerobic pretreatment via two-stage digestion seems to be an efficient biological pretreatment. For landfill, enzymatic treatment may be an interesting solution. Finally, for sludge digestion, both aerobic and anaerobic pretreatments favouring autohydrolysis may be recommended. Full-scale applications already exist but their use remains scarce. Indeed, each biological pretreatment features technological issues. Enzymes have high production costs and limited activity in time. Anaerobic pretreatments, notably two-stage digestion, are more expensive and complex to handle than a single stage. Finally, aerobic pretreatments need fine tuning and control due to respiration mass loss. Research and development conducted toward these specific issues may allow these pretreatments to become more cost-effective as well as practical and thus facilitate their development at fullscale.

1. Introduction

In Europe, France and the United Kingdom have a similar strategy for their biogas industry: a progressive development based both on codigestion of a wide range of substrates and on a limited use of energy crops [1]. Similarly, Germany has shifted, since 2012, from an intense use of energy crops toward a more diverse use of substrates. This new state of affairs in Germany, the biogas European leader, emerged from the quick growth between 2000 and 2012 of an industry that was technologically based on a "standard" liquid CSTR plant using energy crops (especially maize) coupled with cattle liquid manure that permits to ensure high and resilient biogas production [2]. However, this strong appetite for energy crops led to agricultural distortions such as the increase of land rental prices, the increase of energy crops prices (also

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Abbreviations: AD, Anaerobic Digestion; ADT, Advanced Digestion Technology; BMP, Bio-Methane Potential; COD, Chemical Oxygen Demand; CSTR, Continually Stirred Tank Reactor; DM, Dry Matter; DSM, Dutch State Mine; EPS, Extracellular Polymeric Substance; FW, Food Waste; GE, General Electric; HRT, Hydraulic Retention Time; HTP, HydroThermal Pretreatment; IPF, Inverted Phase Fermentation; LCFA, Long Chain Fatty Acid; LFD, Liquid Fraction of Digestate; LiP, Lignin Peroxidase; MCHCA, Microbial Consortium with High Cellulolytic Activity; MnP, Manganese Peroxidase; MSW, Municipal Solid Waste; OFMSW, Organic Fraction of Municipal Waste; OLR, Organic Loading Rate; RPM, Revolution Per Minute; RURAD, RUmen Derived Anaerobic Digestion; s-COD, soluble Chemical Oxygen Demand; SRT, Solids Retention Time; SS-AD, Solid State Anaerobic Digestion; TAD-MAD, Thermophilic Aeerobic Digestion coupled with a Mesophilic Anaerobic Digestion; TMP, Thermophilic Microaerobic Pretreatment; TPAD, Temperature Phased Anaerobic Digestion; TRL, Technological Readiness Level; UASB, Up-flow Anaerobic Sludge Blanket; VFA, Volatile Fatty Acid; VP, Versatile Peroxidase; VS, Volatile Solids; WAS, Waste Activated Sludge; WRF, White Rot Fungi; WWTP, Waste Water Treatment Plant

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used for cattle feeding) and therefore the difficulty for smaller farms or biogas exploitations to thrive [3]. Furthermore, "maizification" (wide spreading of maize monoculture) diminished both beneficial environmental impact of biogas production and public acceptance of the technology [4]. Thus, nowadays in Europe, to handle the higher diversity of substrates required by biogas policies, biogas plant model is shifting from a "standard" to a more versatile conception. Pretreatments are identified as a key tool that requires more investigation [5–7].

From a given organic substrate and in absence of oxygen, anaerobic digestion (AD) is a natural occurring phenomenon. AD can be divided in four different steps happening in the following order: hydrolysis, acidogenesis, acetogenesis and methanogenesis. Depending of the substrate composition and its structure, hydrolysis or methanogenesis can be considered as limiting steps. In the case of complex organic substrates, the hydrolysis step is often the limiting step. Indeed, the production rate, the amount and the variety of hydrolytic enzymes released by hydrolytic microorganisms are often not sufficient to adequately degrade a given substrate [8]. On the other hand, in the case of easily degradable organic substrates, a quick and important acidogenesis step can lead to a rapid acidification of the environment, inhibiting the pH sensitive methanogenesis step [9]. Therefore, pretreatments are needed to handle and reduce substrate limitations toward their optimal use in AD.

At lab scale, evaluation of pretreatments on substrates is mainly carried out using bio-methane potential (BMP) tests. It is a batch method that permits to obtain, from a given substrate, its maximal biogas yield by using a high ratio of substrate to inoculum, a diluted environment and a long incubation time. Curves of BMP tests are usually modelled by using first order kinetic relations but it cannot be used to evaluate kinetics of continuous reactors except if more sophisticated modelling procedure is used [10]. Indeed, at pilot or full-scale the conditions are totally changed. Continuous operation, lower dilution and lower ratio of substrate to inoculum are the main differences. Concerning biogas yield in continuous systems, despite a better adaptation of the inoculum to substrate and the fact that endogenous methane production is included, it is likely that biogas yield will be lower than results of BMP tests due to a lower degradation time. This point concerns both raw and pretreated feedstocks. For pretreatment assessments, BMP tests can also be used to compare kinetic of degradation between raw and pretreated feedstocks via curve shapes [11]. This additional information can be used to forecast pretreatment effect on AD in a continuous system, which takes into account both impacts on methane yield and degradation rate. For low hydraulic retention time, continuous system enhancements on AD rate may prevail over biogas yield enhancements. From that, it can be considered that the most promising BMP results for pretreatments are the one displaying both a positive effect on degradation rate and biogas yield. Ideally, to clearly evaluate the interest of a pretreatment, both BMP and continuous reactor results have to be taken into account.

Four different types of pretreatments can be distinguished: thermal, mechanical, chemical and biological. If the first two ones are already applied at full-scale for a variety of substrates (animal by-products, sludge and lignocellulosic biomass), their high demands in heat or electricity are restricting their benefits [12]. Concerning chemical pretreatments, they are for the moment limited at lab scale due to their cost and their environmental consequences even though some alkaline treatments (lime notably) displayed promising results, especially with lignocellulosic substrates and animal by-products [13]. Finally, like all other pretreatments, biological pretreatments are progressively gaining in interest over time as it can be seen in Web of Science® where "anaerobic digestion" and "biological pretreatment" as topics, display both an increasing scientific production over years and a stable 15% ratio of the total "pretreatment" literature from 2011 (Table 1). This can be due to their potential lower energy consumption and promising results, especially with lignocellulosic materials [12,14].

Table 1

Web of Science® bibliomet	ric study with the topics	"biological pretreatment" or
"pretreatment" and "anaer	obic digestion" (January	2017).

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Period	2009–2010	2011-2012	2013–2014	2015-2016
Number of papers about "biological pretreatment"	54	48	72	136
Number of papers about "pretreatment"	141	324	544	830
Ratio biological/all types of pretreatment	38%	15%	13%	16.5%

Prior to detail biological pretreatments, it is important to clearly distinguish them from another current research topic in AD, which is bioaugmentation [15]. This practice was recently defined as the direct addition of selected strain(s) or mixed cultures to AD to improve the catabolism of refractory compounds such as lignocellulosic materials and to increase the methane yield [16]. Despite the fact that bioaugmentation is targeting similar aims as pretreatments, it will not be addressed further in this review as it cannot be considered as a pretreatment but more as an improved inoculation. However, it can be interesting to combine bioaugmentation with pretreatments due to several advantages of this practice (e.g. environment-friendly, cost effective) [16] and it can be noticed that commercial solutions provided by several companies already exist, such as Bioplus by General Electric Water & Process Technologies (Boston, the USA) or Hycura products (Calgary, Canada) for instance.

Biological pretreatments can be divided in three parts: enzymatic, anaerobic and aerobic. The aim of this review is to give an overview of what is done in this field from lab scale to full-scale in function of the substrate. This review will concern agricultural waste, bio-waste that are gathering food waste and organic fraction of municipal solid waste, municipal solid waste and sewage sludge. Concerning animal by-products, as they generally require thermal pretreatment due to sanitary reasons, literature on the application of biological pretreatment is scarce. For this reason, this feedstock will not be included in this review. Promising and recent research results will be presented, with a special focus on methane yield enhancements. Furthermore, non-exhaustive examples of associated full-scale products sold by companies will be described in order to provide to the reader a first insight in existing industrial applications of such pretreatments.

2. Enzymatic pretreatments

Exogenous enzyme additions during AD in order to improve the hydrolytic step of complex organic substrates have been investigated with a growing interest since the mid-1980s. From this literature, it is important to distinguish the four ways to practice enzyme addition as shown in Fig. 1: (1) in a dedicated pretreatment vessel (2) directly in the digester vessel (single-stage process) (3) directly in the hydrolysis and acidification vessel (two-stage process) (4) in the recirculated AD leachate. Besides, it is also worthy to underline that to obtain an increase in biogas production via enzyme addition, parameters such as enzyme activity, specificity to the substrate, quantity, temperature, pH and enzyme stability need to be optimized and are often keys parameters in the economic assessment [17]. This section will give an overview of enzymatic pretreatments with a special focus on results of available commercial solutions. It can be already mentioned that Dupont (Wilmington, USA), Novozymes (Bagsvaerd, Denmark) and DSM (Delft, the Netherlands) are the three key players in the enzyme market. Therefore, enzyme products that will be presented are mostly commercialized by these companies.

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