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Pretreatment and co-digestion of wastewater sludge for biogas production: Recent research advances and trends

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ABSTRACT

Currently, sludge is not considered as a waste any more, since it is capable of producing valuable products. Besides land disposal and thermochemical processes (i.e. pyrolysis, combustion, gasification), biological processes appear as promising valorisation routes to treat wastewater sludge efficiently. Anaerobic digestion (AD) processes are already being applied at industrial scales for the effective disposal and valorisation of sludge. However, methane yields from sludge anaerobic digestion remain low compared to other types of organic waste. Thus, pretreatment and co-digestion contribute to improve the degradability of organic matter and methane potential of sludge, respectively. This paper reviews the recent achievements in sludge pretreatment and co-digestion with other substrates such as the organic fraction of municipal solid waste, fatty waste, lignocellulosic and algal biomass. Furthermore, recent studies combining co-digestion and pretreatment are examined. The paper also provides recommendations to better manage sludge recovery by taking into account multiple aspects such as techno-economic feasibility, the effect of pretreatment on both the physico-chemical properties of sludge and the quality of digestate. The socio-environmental and legislative aspects are also essential in order to ensure the sustainability of the process.

1. Introduction

Sludge is solid waste generated from wastewater treatment operations. Its volumes are currently increasing, especially in urban areas, along with population growth [1]. According to the wastewater treatment process, sludge can be classified into primary, secondary or mixed sludge, which have various physical and biochemical properties (Fig. 1). The first operation in wastewater treatment involves screening to remove large constituents. After the grit and other heavy solids contained in the stream are removed within the grit chamber, the wastewater is conveyed towards the primary clarification step where physical and chemical treatment (coagulation, flocculation, flotation) and sedimentation take place (Fig. 1). Primary clarification removes about 40–50% of solids in wastewater, thus generating 'primary sludge'. Contaminants present in wastewater are removed in the aeration tank where heterotrophic bacteria consume organic matter and nutrients, with oxygen as final electron acceptor, to grow and produce adenosine triphosphate. Microorganisms mixed with solids then settle in the secondary settler. The produced waste, called 'secondary or activated sludge' is partially recycled to the aeration basin (Fig. 1). Supplementary processes such as tertiary treatment could be necessary for residual suspended solids and nutrients (such as nitrogen and phosphorous) to be eliminated. In certain cases, the stream can also be disinfected by UV, chlorine or ozone [2]. The sludge is thickened, dewatered and finally disposed of. Chemical properties of sludge such as pH value, nutrient and heavy metal concentrations as well as the presence of pathogens should be examined before land application [3]. A selection of sludge characteristics, reported in the literature, is presented in Table 1. A high organic matter content clearly indicates the feasibility of biological degradation by composting or anaerobic digestion. Generally, the degradation of primary sludge is easier than for secondary sludge due to their respective compositions. Indeed, primary sludge contains more easily degradable polysaccharides and fats whereas secondary sludge is mainly composed of microorganisms, exopolymeric substances (proteins and polysaccharides), and undegraded organic and mineral matter [4,5].

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Abbreviations

AcoD	Anaerobic co-digestion
AD	Anaerobic digestion
BMP	Biochemical methane potential
BES	Bioelectrochemical system
COD	Chemical oxygen demand
CFU	Colony forming unit
FOG	Fats, oil and grease
FA	Free-ammonia
GHG	Greenhouse gases
HHV	Higher heating values
HRT	Hydraulic retention time
HTC	Hydrothermal carbonization
MFC	Microbial fuel cells
OFMSW	Organic fraction of municipal solid wastes
OLR	Organic loading rate
PHA	Polyhydroxyalkanoates
PS	Primary sludge
SS	Sewage sludge
SCFA	Short chain fatty acids
TPAD	Thermal phased anaerobic digestion
TS	Total solids
TSS	Total suspended solids
US	Ultrasound
VFA	Volatile fatty acids
VS	Volatile solids
VSS	Volatile suspended solids
WAS	Waste activated sludge
WWTP	Wastewater treatment plant

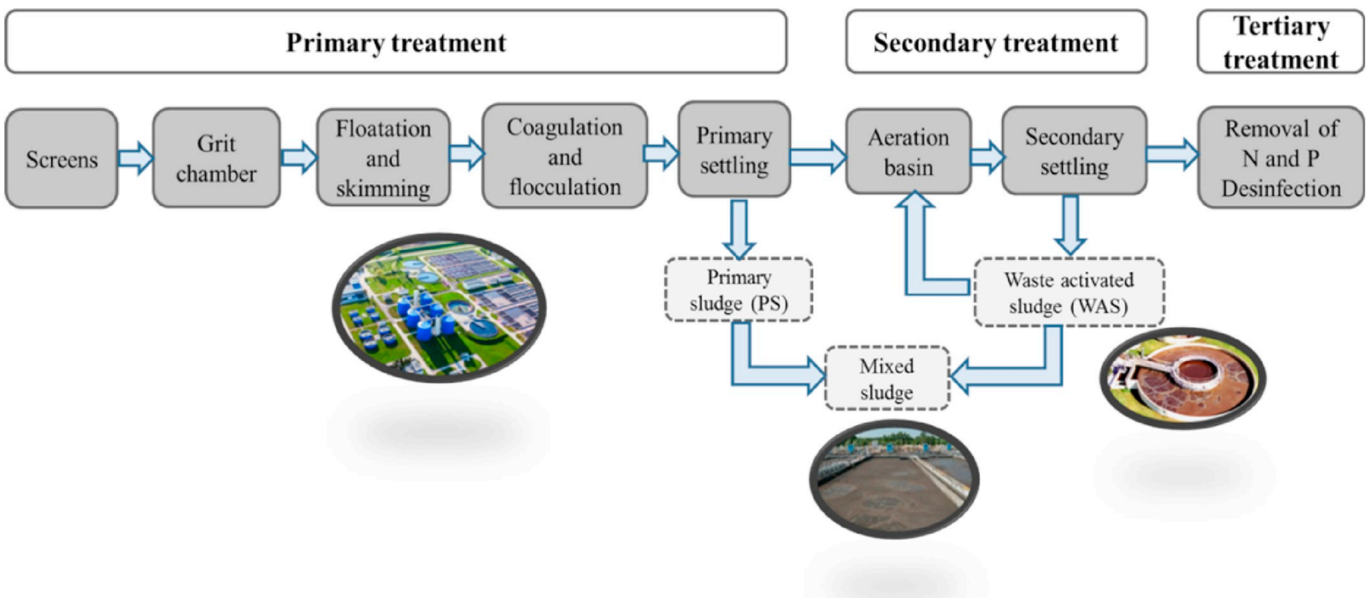

Fig. 1. Wastewater treatment plant processes and classification of sludge.

The management of sludge is complex due to its heterogeneity and potential toxicity [11]. Sludge was formerly considered as waste which could be disposed of by combustion, land application, landfilling and discharges into surface water [12]. These two latter solutions are now forbidden or restricted in most European countries. Combustion reduces the sludge volume while producing energy [13]. Land application is also still widely used as it allows mineral recovery (N, P, K) as fertilizer. It can be preceded by composting, which is a biological stabilization process, allowing for material recovery in agriculture. Anaerobic digestion has been applied for decades in order to reduce both sludge volume and sludge disposal costs. This process has the advantage to produce i) biogas which can be used as an energy source and contributes to wastewater treatment plant self-sufficiency and ii) a digestate which allows, according to the quality of the sludge and to current legislation, nutrient recovery for agriculture [14]. Within the future circular economy, wastewater treatment plants should convert to water-resource-recovery facilities aiming at the efficient reuse of both carbon and nutrients from sludge or from waste water itself. Hence, in the future, sludge should be considered as feedstock for renewable energy and materials [15] as well as for nutrient recovery through agricultural application [16]. For this purpose, emerging biological technologies such as dark fermentation for hydrogen production, bioelectrochemical systems and fermentation for bioplastic production are currently under study. Sludge can also be valorised by thermochemical processes involving gasification [17], pyrolysis [18] and hydrothermal carbonization [19]; they produce energy and biomaterials such as biochars which could be used in agricultural fields and as adsorption materials.

Anaerobic digestion (AD), an environmentally friendly process, is expected to gain a significant role within sludge recovery processes. During AD process, some components of sludge are hardly degraded by microorganisms due to their recalcitrance or to the low accessibility of intracellular matter. A pretreatment process is therefore recommended in order to: i) modify the structure and architecture of the sludge, ii) solubilise organic matter, and iii) increase the accessible surface area and accelerate hydrolysis (the rate-limiting step in the case of secondary or mixed sludge) and consequently improve methane production. As a result, various pretreatment methods have been developed to improve the conversion of sludge into accessible and soluble organic matter in order to maximise biofuel production. Pretreatment technologies include individual or combined mechanical, chemical, physicochemical and biological methods. In recent years, anaerobic co-digestion of sludge (AcoD) was developed with the objective to adjust the carbon/nitrogen ratio, reduce inhibitor concentrations and increase the methane yield. The mixing of sludge with biowaste is restricted in EU countries as mentioned in article 22 of the Directive 2008/98/EC [20].

However, this still remains a feasible option in other countries such as China [21] and India [22]. More recently, combined AcoD and pretreatment of sludge have been investigated [23], although publications on this subject still remain scarce. The efficiency of coupling AcoD and pretreatment depends on the nature and severity of the pretreatment as well as the co-substrate composition. The greatest challenge is yet to choose the right substrate and optimal pretreatment conditions in order to improve methane production and thus better cover the digester energy consumption. Improving the quality of digestate would also allow for its easier handling and management.

The present study first attempts to situate anaerobic digestion among other sludge valorisation processes and reviews recent work on different sludge pretreatments and co-digestion. The novelty of this review lies in its critical analysis of the coupling between AcoD and pretreatment. Furthermore, for the first time, a multi-criteria schematization of sludge valorisation aspects is provided, based on recent research developments and upcoming trends.

2. Sludge disposal and valorisation methods

Fig. 2 illustrates the sludge valorisation pathways reported in this paper. In the first part, classical disposal routes (dumping, landfilling) and valorisation methods (land application and composting) are discussed. A section is dedicated to sludge recovery by anaerobic digestion. Finally emerging biological and thermochemical processes for sludge recovery are assessed.

2.1. Classical sludge disposal and valorisation routes

Although the dumping of sludge into seawater used to be regarded as an economical and acceptable disposal route in the 1980s [24], heavy metal and sediment accumulation in dumping areas finally turned out to be harmful for the local fauna. This practice was therefore restrained thanks to the London protocol in 1996, although more than 80% of sludge generated in China is still dumped into the marine environment [25]. Similarly, landfilling is not a sustainable solution to manage sludge. It generates GHG emissions and leachate to soil or water, which, in the long run, can affect climate change, human health and ecosystem equilibrium through the contamination of plants that can in turn be ingested by fauna. Since 2002, the European Union (EU) legislation has restricted landfilling to ultimate waste. Sewage sludge with higher heating values (HHV) greater than 6 MJ/kg has also been prohibited for landfilling [26] thus opening the way for other disposal routes. Nowadays, the most commonly used sludge management strategies in EU countries include incineration and land application according to the European commission environmental guidelines [27].

Table 1
Characteristics of sludge from literature: Range and reported values of parameters [2,6–10].

Parameters	Primary sludge	waste activated sludge	Digested sludge
TS (%)	2–8	0.83–1.16	6.0–12.0
VS (% TS)	60–80	59–88	30–60
Total COD (g COD/gTS)	1.8–2	1.1–1.4	–
Total COD (g COD/L)	36–144	10.4–15.1	–
TKN (%)	2–5	2–5	2–4
pH	5–8	6.5–8	6.5–7.5
C (%TS)	51.5	53	49
H (%TS)	7	6.7	7.7
O (%TS)	35.5	33	35
N (%TS)	1.5–4	2.4–5	1.6–6
P(%TS)	0.8–2.8	2.8–11	1.5–4
K(%TS)	0–1	0.5–0.7	0–3
S(%TS)	1.5	1	2.1
C/N	13–34	11–22	8–31
Al(%TS)		0.1–13.5	
Ca(%TS)		0.1–25	
Fe (μg/gTS)		1000–154 000	
Mg (μg/gTS)		300-20 000	
Ni (μg/gTS)		2–5300	
Mn (μg/gTS)		32–9870	
Cr (μg/gTS)		10–990 000	
Cd (μg/gTS)		1–3410	
Cu (μg/gTS)		84–17 000	
Pb (μg/gTS)		13–26 000	
Zn (μg/gTS)		101–49 000	
Energy (MJ/kgTS)	23–29	16–23	9–13

Incineration or combustion is a thermal process carried out under excess oxygen conditions. It requires prior or on-site dewatering and drying, but has several advantages: (i) a strong reduction in sludge volume generating stabilized ash, which accounts for only 30% of the volume of dried sludge [28], (ii) it allows the thermal destruction of pathogenic microorganisms as well as (iii) the recovery of energy. Indeed, the calorific value of dry sludge is similar to the HHV of lignite (about 16 300 kJ/kg) [25], which gives sludge the potential as a renewable source of energy especially in countries that suffer from the lack of space for landfilling. Due to the necessity for sludge dewatering or even drying before combustion, the application of incineration is economically expensive. The environmental impact of sludge incineration can be reduced by designing of an efficient energy recovery system, managing fly ashes as well as reducing GHG emissions.

Sludge from wastewater treatment plants contains N, P, K and organic carbon which are essential for plant growth and improving soil properties. Recently, the European Nitrate Directive (Council Directive 91/676/EEC) has fixed a nitrogen spreading load of 170 kg N/ha for vulnerable zones where manure production is in excess [29,30]. The amount of sludge added to commercial crops is generally based on this value. If an average concentration of 35 g_N kg_{TS}^{-1} is considered (Table 1), 6.5 $t_{TSsludge}$ ha^{-1} can be applied. When their concentrations are too low, fertilisation can be completed by mineral P or K. In general, sludge contains both mineral N (NH_4^+) and organic N. As NH_4^+ is more rapidly available, sludge provides less available nutrients than mineral fertilizers [31]. In a study on *N*-mineralization of 12 sewage sludges in a soil during a 16-week aerobic incubation, Serna and Pomares reported that the mineralized N ranged from 13.8 to 45.6% of total organic N applied to soil [32]. Land application is therefore a noteworthy disposal route where nutrient recovery is possible. Nevertheless, the heavy metal content in sludge should be monitored, because of their negative effects on soil. Their accumulation presents potential risks on plant genetics, causing DNA damage. These risks have been ranked in the following order: $As^{3+} > Pb^{2+} > Cd^{2+} > Zn^{2+} > Cu^{2+}$ [33]. Sludge may, in addition, introduce pathogens to the soil (mainly *Salmonella* spp, *Escherichia Coli*, protozoa, viruses and Helminth worms), which have to be removed before any agricultural use of sludge [34,35]. Sludge can also contain organic micro-pollutants [36] such as polycyclic aromatic hydrocarbons [37] and antibiotics [38] which present potential risks for the environment and subsequently human health.

Prior to land application, sludge can be subjected to composting. Composting is a biological process that is widely applied to reduce sludge volumes and effectively minimize their harmful impact on the environment. It is defined as the aerobic stabilization of organic substrates, under conditions that allow an increase in temperature, resulting from biologically produced heat, to produce a compost that is stable, free of pathogens, and that can be beneficially spread on land [39]. It generally concerns solid and semisolid materials. Indeed, compost presents many advantages when applied to land: it helps to reduce pathogens and enhances plant immunity against diseases [40]. Sludge moisture, C/N ratio and aeration conditions can be adjusted by adding another substrate such as green waste, sawdust, straw, corn or rice hulks [41,42].

Finally, from an economic point of view, land application in the European Union is the most cost-effective solution, when compared with other management routes such as incineration, composting and

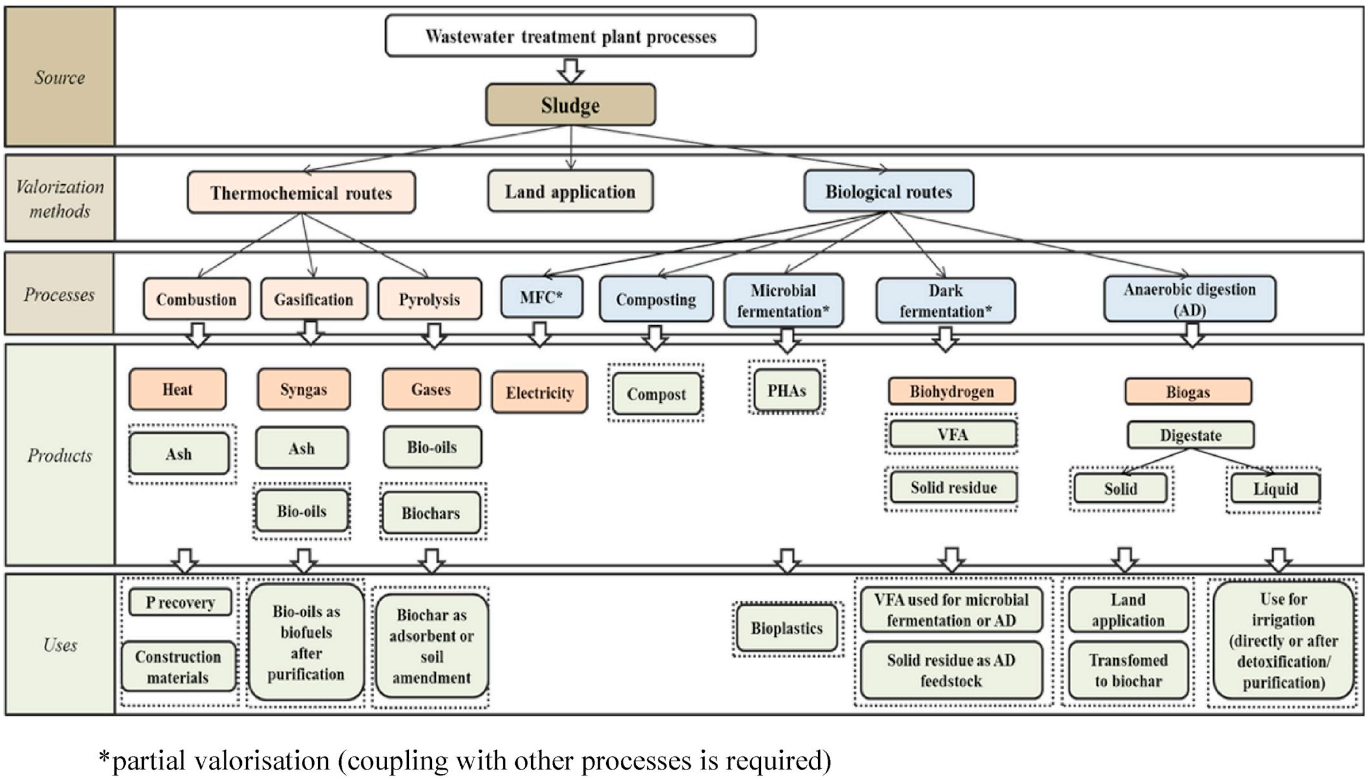

Fig. 2. Sludge valorisation routes reported in this review.

landfilling [43]. Anaerobic digestion which is also a widely spread biological process is described in the next section.

2.2. Anaerobic digestion

The anaerobic digestion (AD) process represents one of the most mature and economically profitable routes towards sludge valorisation [44]. It involves four degradation steps: hydrolysis, acidogenesis, acetogenesis and methanogenesis. In a first step called hydrolysis, large organic polymers (carbohydrates, lipids and proteins) are transformed into monomeric compounds (sugars, fatty acids, amino acids) by strains of hydrolytic bacteria (e.g. Pseudomonas sp.) through extracellular enzymes such as cellulase, amylase, protease and lipase. This is followed by acidogenesis where monomers are degraded into volatile fatty acids such as acetic, butyric, propionic, and valeric acids. Achieved thanks to acidogens (e.g. *Clostridium*, *Ruminococcus*, *Bacillus*, *Escherichia*, Bacteroïdes, *Enterobacter*), acidogenesis is faster than hydrolysis in the case of complex material such as sludge. After acidogenesis, two groups of acetogens coexist; the dominant group (syntrophic acetogenic bacteria) produces acetate, CO_2 and H_2 from volatile fatty acids, while the other, involving homoacetogens, converts CO_2 and H_2 to acetate. Homoacetogens do not play a major role in the AD process, since hydrogen is mainly consumed by hydrogenotrophic methanogens. In the final metabolic stage, methanogens can also be divided into two groups, acetoclastic methanogens which produce methane and CO_2 from acetate and hydrogenotrophic methanogens converting H_2 and CO_2 to CH_4. Theoretically, 70% methane is produced from acetate and 30% from CO_2 and H_2. Thus, the ratio between acetoclastic and hydrogenotrophic communities is 70/30 [45], although this ratio mainly depends on operating conditions, feedstock composition and the presence of inhibitors [46].

The main factors affecting AD performance are pH, temperature, C/N ratio, organic loading rate and mixing conditions. 1) pH should be maintained between 6.5 and 7.5 to balance microbial populations and promote the growth of methanogens. 2) Thermophilic temperature (55 °C) has been found to give the best biogas production rate [4]. However, maintaining this temperature constant requires more energy consumption, while thermophilic digestion is harder to control [47]. Mesophilic temperatures (20–45 °C) are most often used, the optimum being around 33–40 °C. However, as for microbial communities, the effectiveness of mesophilic AD is sensitive to the feedstock composition and operating conditions. 3) C/N ratio depends on the AD substrate and, generally, should vary between 20 and 25. Microorganisms use carbon to grow and nitrogen to build cell structure. However, the C/N ratio of secondary sludge, which is rich in proteins, may be lower than the optimal value (Table 1), leading to risks of ammonia inhibition, especially for thermophilic AD. This risk can be reduced by decreasing both C and N concentrations in the digester. In contrast, certain primary sludges can also present higher C/N ratios than the optimum range (Table 1), especially those that are rich in lipids. Both these limitations can be balanced for mixed sludge. 4) The organic loading rate is defined as the amount of raw material fed per day and per unit volume of the digester. When the anaerobic digestion rate is limited by methanogenesis kinetics, a too high loading rate can entail the accumulation of VFA as well as low methane yields. When the digestion rate is limited by the hydrolysis step as is generally observed for secondary sludge, too high loading rates also result in low methane yields as part of the sludge will remain un-hydrolysed. This parameter therefore needs to be controlled. 5) Mixing conditions should be optimized to maximise the contact between substrates and microorganisms.

Generally, biogas is composed of 50–75% methane and 25–50% carbon dioxide. Biogas can be converted to electrical energy and heat through a combined heat and power unit (CHP). Electricity is injected into the grid while heat is recovered for digester consumption and other applications such as digestate drying. Biogas upgrading is also an option, consisting of methane purification and injection into a gas grid for domestic use and transport. CO_2 and H_2S can be removed using water and amine scrubbers, pressure swing adsorption, cryogenic distillation or membrane gas permeation [48]. Emerging strategies comprise *in-situ* methane enrichment with the addition of activated biochar as adsorbents (CO_2 and H_2S removal) [49], methanation (addition of H_2 to react with CO_2 and produce CH_4) and biological processes using CO_2-fixing microalgae (e.g. *lla minutissima*) [50] or sulfur-reducing bacteria (e.g. *Cholorobium limicola*) [51]. Finally, bioelectrochemical systems (see section 2.3.1) can also be used for the *ex-situ* upgrading of biogas through the electromethanogenesis process. This process consists in the reduction of CO_2 into CH_4 due to the direct transfer of electrons from the cathode to methanogens [52].

Besides biogas, AD also generates a residue called digestate. Provided it does not contain heavy metals, pathogens and/or contaminants, digestate from sludge is generally used as an organo-mineral fertilizer substituting mineral fertilizers, due to its noteworthy content in macronutrients (nitrogen, phosphorous and potassium). For a better management of digestate, the partitioning of nutrients between the liquid and solid fractions is often operated. The separation of digestate can be carried out using centrifuges, screw press, filter press or rotary screens (with or without polymers). Generally, the nutrient-rich liquid digestate (mainly in N and K) is applied as fertilizer while the organic matter-rich solid fraction is used as amender [53].

2.3. Other disposal and valorisation routes

2.3.1. Biological processes: hydrogen, electricity and PHA production

Dark fermentation: Biohydrogen, which is an intermediate product of anaerobic digestion, has a higher value than biogas and represents a promising source of clean energy, as its conversion only generates water without GHG emissions. In order to avoid methanogenic activity, anaerobic digestion process parameters (pretreatment of inoculum, short hydraulic retention time and acidic pH) can be adjusted. The process is thus called dark fermentation and leads to the production of hydrogen and soluble metabolites such as volatile fatty acids and alcohols [54]. However, the hydrogen yield of sludge is very low and few studies have investigated its improvement by the pretreatment of substrate. For example calcium peroxide and nitrous acid pretreatments have led to an increase in the waste activated sludge yield from 0.77 to 10.55 mL/gVSS [55] and from 8.5 to 15 mL/gVSS) [56], respectively. Hydrogen production may be improved by the co-fermentation of sludge with other substrates that enhances the low C/N ratio of the sludge and favours higher amounts of fermentable sugars [54,57].

Generally, as only a small part of the organic load is converted into hydrogen through the dark fermentation process (less than 6% for municipal waste including sludge), dark fermentation should be combined with other processes in order to increase the recovery yields [54]. For example, methane can be produced by anaerobic digestion of both VFA-rich effluents and undegraded solids.

Bioelectrochemical systems (BES) are electrochemical processes in which the oxidation reaction at the anode and/or reduction at the cathode is catalysed by microorganisms. These innovative technologies are used for electrical energy or chemical production and also for environmental services such as water desalination. They can be classified as microbial fuel cells (MFC), microbial electrolysis cell (MEC), microbial electrosynthesis (MES), microbial desalination cell (MDC) and microbial solar cell (MSC). MFC and MEC are the most studied BES. The MFC processes generate electrical current [58], but they are very sensitive to operational conditions which are typically ambient temperature, atmospheric pressure and neutral pH [59]. In addition, MFC are emerging techniques in wastewater treatment, as they reduce the volume of produced sludge, improve nitrogen removal and increase its filtrability for easy handling [60]. Sludge can be used for MFC both as substrate and inoculum. However, MFC is not only used for generating electricity from sewage sludge but also for enhancing the removal of ammonia and organic compounds via COD solubilisation [61]. Indeed, MFC integration in wastewater sludge biological treatment has been studied in lab-scale studies during which MFC was applied to sludge originating from a membrane bioreactor; as a result of the study, the sludge COD and volatile suspended solids were highly reduced [62].

MEC is based on the coupling of organic compound oxidation leading to CO_2 production in the anodic compartment and hydrogen production at the cathodic compartment.

$$2H^+ + 2\ e^- \rightarrow H_2$$

According to Gajaraj et al. (2017), sludge as a MEC subtrate is not relevant for pratical use because it requires high voltage (> 1.4 V) and an addition of mineral media [63]. In different lab-scale studies, MEC was incorporated into anaerobic digesters. Both an enhanced hydrogen production (equation 1) [64] and an enhanced methanogenisis through electromethanogenisis related to the direct electron transfer between the electrode and the cathodic film (equation 2) [65] were reported.

$$CO_2 + 8H^+ + 2\ e^- \rightarrow CH_4 + 2H_2O$$

Results include the sludge retention time that could be shortened from 15 days to 6 days while an efficient sludge reduction could be maintained [64]; in addition, the methane production could be increased by about 8% at low voltage (0.3 V) [63].

The use of BES on digestates from the AD process has been suggested for effluent polishing, removing residual soluble COD while producing electricity [52,59]. MEC can also be applied to the VFA-rich effluent from dark fermentation, in order to increase hydrogen production [59].

Research on BES is currently very active but the scalability and implementation of such processes are still very uncertain [52]. Furthermore, both dark fermentation and BES allow partial recovery of the sludge stream and cannot be considered as final disposal routes. In consequence, they should be combined, eventually with each other, but especially with other processes such as anaerobic digestion, that would promote sludge volume reduction and further recovery.

Polyhydroxyalkanoates (PHA) are highly biodegradable polymers that can substitute synthetic plastics. They are produced by fermentation as a form of energy storage in mixed consortia of microorganisms, which are generally selected from activated sludge. They are fed with VFA-rich streams (that can originate from acidogenic fermentation of primary or secondary sludge). PHA accumulation in bacteria is favoured by nutrient starvation and by a feast/famine strategy alternating carbon excess and limitation. PHA then are extracted by a solvent addition, flotation or digestion method [60]. PHA production from sludge has been carried out at lab and pilot scales [61], as well as full scale [66,67]. This process also has the advantage of minimizing sludge volume with a COD removal attaining 89% [68]. Although the extraction methods of these biopolymers have recently been developed [69], they are still expensive [70].

2.3.2. Thermochemical processes for energy and biochar production

Gasification is employed for converting feedstock into fuel gases and chars, by supplying less oxygen than the stoichiometric requirement. The most noteworthy advantage of this process is the low quantity of generated solids, which minimises the issue of their disposal. However, N, S, Cl and F are converted to NH_3, H_2S, HCl and HF, respectively. These compounds should be controlled and trapped because of their undesirable effects on the environment [71]. Indeed, the main issues related to the gasification process are: the production of toxic gases, fire risks, explosion risks and soil contamination by ashes and tars.

Pyrolysis is the conversion of waste biomass into bioproducts and energy in the form of gases, oils and chars. It takes place via the thermal degradation of a complex material in an inert atmosphere or vacuum [72]. Gaseous effluents and bio-oils can be used for generating energy, while biochars can have a role as adsorbents of heavy metals [73-75] or soil amendments [33,76,77] depending on the quality of the char that is defined by its ash content [78,79]. The energy consumption by pyrolysis can be reduced with a microwave-assisted pyrolysis device. The latter was studied in recent work [80,81] during which the resulting bio-oils had higher yields when compared to conventional pyrolysis and shared similar properties to biodiesels [82]. When the feedstock contains high amounts of water, it has to be either dried before pyrolysis or directly subjected to a wet pyrolysis operation called 'hydrothermal carbonization'.

Hydrothermal carbonization (HTC) has been defined as an exothermic reaction that converts organic waste or biomass into a solid coal-like substance, thus imitating the natural coalification process. The main advantage of this process is that it does not require any drying step, which is particularly suitable for sludge that does not need dewatering. In addition, due to its high nutrient content, the produced hydrochar can be utilised as a soil amendment, thus increasing carbon storage in the soil. Even though biochars from pyrolysis and hydrochars are used in similar ways, the latter decomposes more easily (less stable in soil) than biochars due to their weaker aromatic structure and higher percentage in labile carbon species [83].

Besides the generation of valuable products, all these thermochemical processes can be carried out as sludge disposal methods since they significantly reduce sludge volumes.

Table 2 summarizes the valorisation and disposal processes cited in this review and shows their benefits, drawbacks and recommendations.

3. Sludge pretreatment to enhance methane production

Pretreatment of sludge prior to biological conversion were investigated in a vast number of papers that have been reviewed by different authors [23,93–96]. Table 3 summarizes recent pretreatment methods developed for sludge. The pretreatments are needed to increase methane production by improving the biodegradability of organic materials, and their accessibility to microorganisms. Indeed, pretreatments aim at: i) improving the digestibility of organic matter, ii) increasing the hydrolysis rate, iii) improving the methane yield, iv) enhancing dewaterability and v) reducing sludge viscosity (decreases pumping costs). Pretreatments can be organised into different categories: physical, chemical, biological, thermal or combined pretreatments.

3.1. Physical pretreatments

Different physical pretreatments have been developed and reported in literature for sludge disintegration such as grinding, pulse electric fields, high-pressure homogenization, lysis centrifuges, microwave irradiation and ultrasonication.

Grinding reduces particle sizes and breaks up flocs, it enhances sludge disintegration which then accelerates its AD [110]. The soluble COD and methane production in anaerobically digested sludge after grinding have been found to be higher than for waste activated sludge. This could be related to the lower biodegradability of digested sludge [23].

Pulsed electric fields lead to sudden disruption of cell walls and solubilisation of macromolecules and complex organic matter under high voltage [93]. Focused-pulsed pretreatment was experimented on waste activated sludge under 34 kWh/m^3 and a SRT of 20 days, resulting in an increase in soluble COD, soluble sugars and proteins by 220%, 300% and 460% respectively. Consequently, methane production rose by 33% [111]. By applying an energy of 10 kWh/m^3, Salerno et al. [112] found that methane production from waste activated sludge doubled after 25–30 days of biochemical methane potential (BMP) tests. This technology was applied to mixed sludge (60% primary + 40% waste activated sludge) under 16 kWh/m^3. The obtained methane yield was 30% higher than for untreated sludge [113].

Table 2
Valorisation and disposal processes of sludge: advantages, disadvantages and recommandations

Process	Advantages	Disadvantages	Recommendations	Ref
Combustion	Significant reduction of sludge volumes Energy recovery Destruction of pathogens Mature technology	High energy consumption Drying/dewatering is required	Management of ashes (concentrated in heavy metals) is required. Optimization of energy consumption Gaseous emissions treatment	[28]
Gasification	Sludge volumes reduction Energy production (syngas)	Complexity of the technology High energy consumption Drying/dewatering is required	Optimization of operating conditions Co-gasification with other wastes to reduce dewatering costs Gaseous emissions treatment	[2]
Pyrolysis and HTC	Sludge volumes reduction Energy production (syngas and bio-oils) Biochars generation (adsorption properties depending on operating conditions)	Complexity of the technology High energy consumption and investment costs Drying requirement (in the case of pyrolysis)	Optimization of operating conditions Gaseous emissions treatment Co-pyrolysis with more carbonaceous materials	[28]
BES	Reduction of COD and ammonia in sludge and pollutants removal Electrical energy generation Hydrogen production	Expensive scale-up Low power generated	Coupling MFC with other processes such as anaerobic digestion to enhance its effectiveness Optimization of electrode materials selection	[84,85]
PHA production	Reduction of sludge Bioplastics production	Expensive technology Wastes disposal costs	Pretreatment of the feedstock Development of low-cost extraction methods Coupling the process with AD or composting for an effective disposal of the remained solids	[86]
Dark fermentation	Low energy consumption Biohydrogen production	Low yields of biohydrogen Pretreatments of feedstock may be needed Sustainable management of the residues is required	Optimization of pretreatment conditions PHA rich effluent must be valorised (AD for example)	[87,88]
Composting	Reduction of sludge volume Stabilization of sludge Degradation of pollutants Low energy consumption Cost effective in large scales Mature technology	Dewatering of sludge may be needed.	Co-composting of sludge with other organic wastes may enhance the compost quality.	[89,90]
Anaerobic digestion	Reduction of sludge volume Energy production (biogas) Low energy consumption Digestate used as organic fertilizer mature technology	Low degradability of some sludge which may require pretreatments (additional costs) Digestate management strategy required	Optimization of biogas upgrading techniques to reduce the process costs Optimization of pretreatments conditions If not suitable for agricultural use, digestate can be subjected to thermochemical processes	[23,91,92]

Table 3
Pretreatments in literature (2017–2018).

Substrates	Pretreatments	Conditions	Effects	Ref
Sewage sludge	Thermal hydrolysis	180 °C for 76 min	+340% of methane produced	[97]
Waste activated sludge	Bio-electro Fenton		+32% of methane produced	[98]
Sewage sludge	Microwaves	20 000 J/g TS for 63s	+10% of soluble COD At OLR = 2.6 gVS/L.d, +20% methane production	[99]
Sewage sludge	Ultrasonic	15 min in an ice bath, 20 kHz, 50 W (353 J/gTS)	+34% of methane produced	[100]
	Thermal hydrolysis	30 min, under 2 bar and 120 °C in autoclave	+51% of methane produced	
Waste activated sludge	Free-ammonia and heat pretreatment	135.4 mg FA/L at 70 °C 24 h	+25% of methane produced	[101]
Waste activated sludge	Microwave and H_2O_2 pretreatment	600 W until 80 °C. 0.2 g H_2O_2/g TS Heating rate of 20 °C/min	+20% of methane produced Decrease the viscosity of sludge	[102]
Waste activated sludge	Calcium peroxide and free ammonia pretreatment	0.05 g/g VSS of CaO_2 180 mg/L of FA for 3 d	Higher soluble COD (6-fold the sCOD from untreated sludge) Increase SCFAs from 25 to 350 mg/L	[103]
Thickened mixed sludge	Ultrasonic and low-temperature pretreatment	30 500 kJ/kgTS (26 kHz) 55 °C at 70 rpm for 13 h	+50% of methane produced Negative energy balance (−73 MWh/d)	[104]
Thickened activated sludge	Radio frequency heating system	Radio frequency 13.56 MHz 120 °C for 2 h	Soluble COD increased (5 fold the sCOD of untreated) +16% of biogas produced	[105]
	Microwaves	Microwaves frequency 2.45 GHz 120 °C for 2 h	Soluble COD increased (5 fold the sCOD of untreated) +14% of biogas produced	
Waste activated sludge	Ultrasonic	35 000J/gTS 20 KHz 34 min	+31% of methane than untreated	[106]
Waste activated sludge	Surfactant coupled sonic pretreatment	Dioctyl sodium sulphosuccinate 0.05 g/g suspended solids Ultrasonic 5120 J/g TS	+20% sludge lysis rate Soluble COD is 10-fold the sCOD of untreated sludge	[107]
Secondary sludge	Biological pretreatment	42 °C under 100 rpm for 6 days Anaerobic conditions	Same methane volume was produced after 15 days, compared to untreated sludge after 30 days of BMP (enhanced kinetics). 3-log reduction of *E. coli* +33% volatile solids reduction	[108]
Dewatered sewage sludge	Ca $(OH)_2$ and ultrasonic pretreatment	0.04 g of lime per gTS for 1 h 225 kJ/kg TS for 15 min	sCOD 4 times higher than untreated sludge +60% of methane produced	[109]

However, the use of focused-pulsed pretreatment on primary sludge only slightly increased the methane production (by 8%) [114]. The impact of this physical method on primary sludge is therefore weaker than the impact on waste activated sludge. A full-scale trial was carried out implementing pulsed electric field technology prior to two 3300 m^3 digesters. The energy balance showed that treating 380 m^3 per day of mixed sludge should generate at least 2.7 fold energy benefits and as much as 18 fold the pretreatment energy input, depending on the heat recovery. The payback period was estimated to 3 years [115].

High-pressure homogenization (up to 900 bar) is another method for cell disruption and floc disintegration. It was first used for food and dairy emulsions, and then applied to wastewater sludge [116]. Methane production increased by 60% after homogenization under a pressure of 300 bar [117]. Wahidunnabi et al. [118]demonstrated that high-pressure homogenization under 827 bar increased sugar and protein solubilisation from almost 2%–15% and when combined with two-phased anaerobic digestion. Statistically, a significant improvement of 81% of methane production was observed while the digester volume was reduced by 33%. The energy balance was positive, covering the total pretreatment energy requirements.

Lysis centrifuge consists of a simple device adapted to a centrifuge, causing partial destruction of sludge cells [119]. In addition, it can enhance 15–26% of biogas production from thickened sludge [119], without requiring any additional operation, as the centrifuge is also used for sludge dewatering prior to AD.

Microwave irradiation ensures a quick disintegration of sludge, enhances its solubilisation and degradability, and thus increases the hydrolysis rate. Furthermore, it destroys the faecal coliforms and *Salmonella* spp contained in sludge [120], is inexpensive and has a minimal impact on the environment [121]. Park et al. [122]showed that application of microwaves for 7 min with an energy density of 1260 kJ/m^3 resulted in 20% and 29% higher COD solubilisation and methane production respectively, in comparison to untreated sludge, thus confirming the self-sufficiency of the pretreatment. According to Neumann et al. [123] microwave irradiation operating with a specific energy of 336 000 kJ/m^3 and a power value between 800 and 1250 MW resulted in an enhancement of volatile solid reduction from 23% to 48% and an improvement from 16 to 50% biogas production. Also, working with a specific energy of 20 000 kJ.kgTS^{-1}, this process resulted in a 20% rise of methane production, while soluble COD increased by 10%. However, the application of a high energy density requires a higher methane increase in order to cover the pretreatment consumption, as the gained energy only represents 5% of the microwave consumed energy. The power or energy density of microwave treatment as well as the pretreatment temperature need to be optimized in order to ensure pretreatment efficiency [99]. Housseini Koupaie et al. [105] compared the effect of radio frequency systems with microwave pretreatments on sludge disintegration and anaerobic digestion. For the same temperature and pretreatment time, soluble COD after both procedures were similar. Also, the difference in observed methane production was not significant. Nevertheless, 19% more sugars were solubilised after microwave treatment.

Similarly, other types of irradiation such as the **electron beam**, **beta** and **gamma** irradiation have been applied to sludge. Electron beam impacts cross-linked polymers and destroys any form of life in sludge [124]. **Gamma** and **beta radiation** can also disintegrate sludge and facilitate its accessibility [125].

Ultrasonic pretreatment enhances digestion stability, sludge dewaterability, solubilisation of volatile solids and biogas production. It generates a digestate with low residual organic matter [126,127] and low pathogen counts by reducing filamentous organisms [128]. It also modifies the physical characteristics of sludge such as settling velocity, floc structure and particle size which, unlike turbidity, decreases with the supplied energy. Chemical characteristics of sludge also change after the ultrasonication [129]: soluble polysaccharide and protein concentrations increase due to cell disruption and the release of organic macromolecules [130]. Ultrasonic treatment enhanced methane production by 34% at an energy supplied of 14 000 kJ kg TS^{-1}. The methane enhancement can cover 80% of ultrasonication consumption [1] [100]. However, a higher energy supply (35 000 kJ kg TS^{-1}) does not further enhance methane production. According to Carrere et al. [23] energies supplied in the range of 1000 kJ kg TS^{-1} and 16 000 kJ kg TS^{-1} result in higher methane yields by up to 140% of enhancement in batch reactors.

3.2. Thermal pretreatment

Similarly, sludge solubilisation improves after thermal hydrolysis (> 100 °C). Thermal pretreatment was initially used to enhance sludge dewaterability, then it was applied as a pretreatment step for ensuring sludge partial solubilisation and its complete disinfection [131]. The optimal temperatures for sludge thermal pretreatment range between 150 °C and 180 °C under a pressure of 600–2500 kPa and during 30–60 min. Above these temperatures, solubilisation increases while methane production decreases. This can be assigned to Maillard reactions and formation of recalcitrant matter [23]. Bougrier et al. [132] observed that methane obtained from thermally pretreated sludge (at 170 °C) was 51% higher than that from untreated sludge with full coverage of the energy dissipated during pre-treatment.

3.3. Chemical pretreatment

Most studied chemical sludge pretreatments use alkali and acidic reagents which solubilise proteins and sugars respectively [133]. **Alkaline pretreatment** used for sludge includes NaOH, KOH, $Mg(OH)_2$ and $Ca(OH)_2$, with a pH ranging from 8 to 12 for 30 min to 8 days. However, the nature and concentration of chemical reagents should be carefully selected to avoid inhibition. **Free-ammonia** is another chemical reagent that can be used for sludge pretreatment. Wei et al. [134] reported that free-ammonia at pH 10 enhances solubilisation and methane production, while the hydrolysis rate can double. Free-ammonia addition is a noteworthy approach for pretreating sludge before its anaerobic digestion, indeed recovering ammonia from the digestate would make this process more environmentally friendly [134]. However, to avoid inhibition, the removal of ammonia prior to AD of the pretreated sludge is necessary [135]. The most common method for ammonia recovery is striping. Furthermore, recent studies investigated sludge pretreatment using free-ammonia combined with other chemical reagents. A significant increase in the solubilisation and short-chain fatty acid production have been observed after both calcium peroxide and a biosurfactant (rhamnolipid) were combined with free-ammonia [103,136]. Nevertheless, methanogens were highly inhibited.

Oxidation with ozone or H_2O_2 has also been applied to pretreat sludge. **Ozonation** favours cell disruption, and the solubilisation of materials that are difficult to degrade. It also causes the mineralization of organic matter at higher doses. Silvestre et al. [137] reported that the optimal dose of ozone was 0.06 $gO_3.gTSS^{-1}$ which led to a two-fold higher methane production than for untreated sludge while Chacana et al. [138] found that 0.08 $gO_3.gCOD^{-1}$ increased the methane yield by 16%. However, a risk assessment highlighted that ozone doses between 13 $mgO_3.\ g_{sludge}{}^{-1}$ and 38 $mgO_3.\ g_{sludge}{}^{-1}$ can intensify sludge impact on the environment by increasing the acid-soluble/exchangeable fraction of heavy metals in comparison with untreated sludge [139]. **Hydrogen peroxide** is another strong oxidant ($E_0 = 1.76$ V NHE) that has been used for sludge pretreatment. It can react directly with molecules via redox pathways, but this oxidation occurs at high temperatures. Valo et al. [140]reported that hydrogen peroxide increased soluble COD by 30%, although no positive impact could be observed on sludge biodegradability. However, when a catalyst ($FeSO_4$) was added, the pretreatment resulted in an increase of 16% of biogas production. Fenton's reagent is used to solubilise refractory substances and improve sludge dewaterability [141]. A bio-electro-Fenton system has also been studied [98]. The anode and cathode presented different impacts on the cell breaking of Gram-positive and Gram-negative bacteria. Although the highest methane yield resulted from the cathode-treated sludge, the anode was more efficient in macromolecule destruction and sugar solubilisation [98]. Cation-exchange resins can also be employed to adsorb divalent cations in the supernatant; this accelerates hydrolysis and promotes solubilisation of organic substances, thus increasing the methane production rate [142,143].

3.4. Biological pretreatment

Biological pretreatments include thermal phased anaerobic digestion, enzymatic hydrolysis and addition of fungi or bio-surfactants [93]. Thermal phased anaerobic digestion (TPAD) is the most reported biological treatment. It consists in the pre-hydrolysis of sludge before its AD in two stages at different temperatures [144]. Besides the increase in solubilisation and methane production, TPAD energy consumption is quite low. Ding et al. [108]investigated the biological pretreatment of waste activated sludge under anaerobic conditions and at different temperatures. Enhanced hydrolysis rates and removal of volatile solids were obtained at 42 °C and a 3-log reduction of *Escherichia coli* was observed. A thermal pretreatment at 165 °C for 30 min and biological pretreatment at 42 °C for 3 days were found to have a similar biochemical methane potential after 30 days of anaerobic digestion. In another study, the autohydrolysis of WAS at 55 °C for 12–24 h increased methane production by 26% which was quite lower than the thermal pretreatment at 170 °C for 30 min (45% methane increase). However, the energy produced covered the energy needs for AD and autohydrolysis, with an electrical energy gain of 0.3 MW instead of 0.2 MW if sludge had not been treated [145]. Ge et al. [146] reported that a 25% higher methane production in mesophilic anaerobic digestion resulted after a biological pretreatment at 50–70 °C for 2 days. Finally, the highest improvement in methane production was reported by Bolzonella et al. [144], considering that a 69% and 145% higher biogas yield was obtained under pretreatment temperatures of 47 °C and 70 °C respectively for 2 days.

3.5. Combined pretreatments

Combined pretreatments are being widely investigated in order to achieve synergetic impacts on sludge AD. Various pretreatments can be coupled, as for example, ultrasound and Fenton [147], microwave and acidic [148], thermo-alkaline and ultrasound [149]; alkaline and gamma irradiation [150] and thermo-alkaline pretreatments (> 100 °C) [140]. Thermo-alkaline pretreatment is most frequently reported in literature [151,152]. Park et al. [122]described an increase in solubilisation and methane yields up to 87% and 154% respectively in comparison to untreated sludge. The most recent combinations of pretreatments studied in literature are summarized in Table 3. Liu et al. [153] pretreated sludge with free-ammonia under 70 °C. The combined pretreatment resulted in an increase of 25% of methane production compared to untreated sludge. The energy input required for this pretreatment is covered by the energy benefit. Furthermore, surfactants coupled with ultrasonic pretreatment significantly enhanced the solubilisation of sludge, which reached 10 fold the solubilisation of untreated sludge [107]. In another study, the combination of ultrasonic and lime pretreatments resulted in a 60% methane enhancement, achieving higher soluble COD compared to ultrasonication alone. The addition of lime decreased the energy consumed by ultrasonic pretreatment by about 67% (standard deviation 6%) [109]. The combination of oxidation with microwaves [102], ultrasonication [154] and other pretreatments such as hydrodynamic cavitation [155] has also been examined.

4. Discussion

Research on sludge pretreatment methods for their anaerobic properties is very active at lab-scales, with investigations on various techniques and more particularly on certain combined processes. The first objective of such studies is to increase the biogas or methane production. However this criteria strongly depends on many parameters such as i) the nature of sludge (waste activated sludge undergoes better improvements than mixed and primary sludge; among waste activated sludge best results are obtained with those from extended aeration processes which have the lowest intrinsic biodegradability [156]). ii) conditions of anaerobic digestion processes: may be either batch or continuous modes. In continuous AD, the lower the hydraulic retention time, the higher the impact of pretreatment as pretreatment results integrate the enhancement of the digestion rate in addition to the enhancement of the methane yield (BMP). Table 4, which provides an overview and compares the different kinds of pretreatments, thus reports a rough qualitative assessment of their impact on methane production.

The energy requirement of pretreatments is a key parameter. First, pretreatments are sorted according to electrical and heat requirements. Heat energy is more available in wastewater treatment plants than electrical energy. Biogas can be valorised by a combined heat and power (CHP) engine: electricity is usually sold whereas heat remains in excess. Thus heat-consuming pretreatments should be favoured rather than those that consume electricity. Thermal pretreatment or steam explosion operating at 160–170 °C is one of the most implemented techniques at full scale [96]. The impacts of microwave pretreatment resemble those of thermal treatment [157], however the former type of pretreatment requires electrical power and, so far, does not have a full scale application.

In addition, the energy balance of pretreatments, defined as the excess energy produced subsequent to pretreatment minus the energy consumed by the pretreatment step, strongly depends on the sludge TS concentration. This is obvious in the case of thermal treatment, where the lower the sludge TS concentration, the higher the energy that is wasted to heat water. It is thus highly recommended to concentrate sludge before pretreatment. However, a too high concentration can produce adverse effects on sludge rheology and transportability as well as on possible inhibitor concentrations. Cano et al. [96] proposed a linear relationship between the maximum consumed energy and the sludge concentration. Significantly different trends were found between lab- and full-scale pretreatment devices, the full scale systems being more efficient energetically. In addition, heat recovery during thermal pretreatment allowed for a significant reduction in energy requirements. As for electricity-consuming techniques, full-scale sonicators were found to be most energy efficient together with lysing centrifuge whereas lab-scale sonicators were inefficient. These authors report an efficient ultrasonic pretreatment when a typical energy value of 6 kWh/m^3 was consumed [96]. However, this result depends on the TS concentration in sludge. Other full-scale applied techniques were classed at the limit of electrical efficiency (ball mill grinding and pulsed electric fields) or even in the inefficiency domain (high pressure) [96]. Nevertheless, in spite of a negative energy balance, pretreatments can prove to be economically interesting, when the sludge volume and the sludge disposal costs can be reduced.

Biological pretreatments require less energy supply and are generally self-sufficient; even though investment costs can make the process less attractive, two phase or temperature-phased process have been implemented at full-scale.

To our best knowledge, chemical pretreatments prior to sludge AD are not used in waste water treatment plants. When chemical reagents are used, care should be taken that i) the AD process is not inhibited; ii) there is no negative impact on the quality of the digestate as this could in turn limit its return to the soil; iii) pretreatment costs are reduced; iv) the effect of waste generated during chemical reagent production is minimised. Generally, when combined with physical or thermal techniques, chemicals can significantly improve methane production while reducing the energy consumption of the physical or thermal process. Nonetheless, the added costs of both processes do not allow for the full scale application of such processes.

5. Co-digestion of sludge

Sludge can either be digested alone, or mixed with other types of organic waste, in order to enhance biogas production. Co-digestion applied to sludge presents several advantages. For sludge, the methane production can increase if the methane potential of the co-substrate is sufficiently high. It can also equilibrate the C to N ratio. For the co-substrate, co-digestion adjusts the moisture content, C/N ratio, nutrient balance and dilutes toxic compounds, as well as avoids inhibition [135,158,159]. For this reason, the knowledge of the characteristics of each component and their digestion behaviour helps scientists to identify the best 'organic couples' that provide a synergetic digestion performance.

Lipids have the highest methane potential due to the high number of

Table 4
Overview of main sludge pretreatments adapted from Carrere et al. [23] and Cano et al. [96].

Pretreatment	Increase in biogas production	Full Scale	Energy demand	Advantages	Drawbacks
Grinding	+	yes	electrical	Simple	High energy demand
Pulsed fields	+ +	yes	electrical	Low retention time	
High pressure	+ +	yes	electrical	Low retention time	High Capex
Lysing centrifuge	+	yes	electrical	Low cost, Low energy demand	
Microwawe	+ +	no	electrical	Low retention time	High energy demand
Sonication	+ +	yes	electrical	Low exposure time	High energy demand
γ and β radiations	+	no	electrical		High cost
Thermal	+ + +	yes	heat	Sanitation Viscosity reduction	High heat demand Risk of recalcitrant compounds formation High CAPEX
Chemical	+	no	chemical productions	Simple process	Chemical contamination of digestate Risk of inhibitor formation
Thermo chemical	+ + +	no	chemicals productions + heat	Lower energy demand than thermal alone	Chemical contamination of digestate Risk of inhibitor formation
Combined	+ + +	no	electricity + chemical production	Lower energy demand than physical alone	High costs Chemical contamination of digestate Risk of inhibitor formation
Biological	+ +	yes	heat	Low energy demand	High exposure time

carbon and hydrogen atoms in their molecules, although inhibition of methanogenic archaea and foaming problems can occur. Lipids can be found in meat processing by-products, fatty wastewater and some agro-industrial residues such as olive and soybean residues. Carbohydrates are easily biodegradable and well-known for their rapid conversion, but deliver lower methane yields. Carbohydrates are contained in agricultural wastes and in the organic fraction of municipal solid wastes (OFMSW), more specifically food waste. Proteins are essentially found in waste from slaughterhouses and meat processing. They are suitable for co-digestion because of their high organic content; however when digested alone, their high nitrogen concentration can cause process inhibition due to ammonia [160].

Table 5 presents the concentrations of carbohydrates, proteins and lipids contained in different types of waste as well as their experimental BMP. In addition, Table 5 reports the theoretical contribution of carbohydrates, proteins and lipids to the BMP residue, based on the content (% TS) of each component. According to Moller et al. [161], the theoretical methane production from carbohydrates, proteins and lipids is 415, 496 and 1014 mLgVS^{-1} respectively. Table 5 clearly demonstrates that experimental BMP of sludge and agricultural waste are much lower than the theoretical values, due to the low biodegradability and bioaccessibility of certain organic compounds. Rising research interest is given to the co-digestion of sludge. Indeed, the number of articles dedicated to the co-digestion of sludge between 1956 and 2019 is about 242, including 133 articles published since the past 5 years. This review focuses on current studies of co-digestion of sludge between 2013 and 2019 (Fig. 3). The most commonly reported types of waste used for sludge co-digestion are food waste, the organic fraction of municipal solid waste, agro-industrial and fatty waste. In addition, co-digestion with agricultural waste, microalgae and other waste has also been investigated. Results from previous studies are presented in Table 6.

5.1. Organic fraction of municipal solid waste (OFMSW) and food waste

The organic fraction of municipal solid waste comprises food waste, yard waste, paper and newspapers [163]. OFMSW has been widely reported as a co-substrate for sludge due to the high biodegradability of the food waste fraction, and to the high C to N ratio of the paper and bio-waste it contains. Tyagi et al. [163] reviewed the optimal parameters for the AcoD of sludge and OFMSW. Process stability is reported to be ensured when OLR values lie between 1 and 3.5 kg VS.m^{-3}.d^{-1} in lab and pilot scales, and between 0.78 and 3.2 kg VS.m^{-3}.d^{-1} at full scale. As an example of a successful full-scale experiment of AcoD of mixed sludge and OFMSW, Zupancic et al. [186] reported an increase in the methane yield from 0.39 m^3 kg^{-1} volatile suspended solids when the digester was fed by sludge (OLR = 0.8 kg VS.m^{-3}.d^{-1}) to 0.60 m^3 kg^{-1} in the case of AcoD (OLR = 1.0 kg VS.m^{-3}.d^{-1}) with a sludge/OFMSW ratio of 80/20 on a TS basis. Mattioli et al. [187] reported an enhancement in biogas production rate from 0.21 to 0.43 m^3 m^{-3} d^{-1}, when the organic loading rate of a mixture of 60% of sludge and 40% of organic solid waste increased from 0.73 to 1.38 kgVS.m^{-3}.d^{-1}, thus covering 50% of the plant's energetic demand. The co-digestion of 40% OFSWM and 60% sludge was observed to be profitable after using life cycle assessment calculations, especially if the digestate is managed [188]. Furthermore, according to the energetic balance, co-digestion can provide energy for the overall process, considering that the power conversion efficiency of a combined heat and power system is 30%.

Food waste contained in OFMSW was also studied separately as a substrate for sludge co-digestion. Kitchen waste, and fruit and vegetable waste, were considered here as food waste. These have been the most commonly studied substrates for co-digestion with sludge since the past 5 years, as shown in Fig. 3. The AcoD of sludge with food waste improves the methane yield in comparison to sludge monodigestion. This increases along with the fraction of food waste which has a higher methane potential than sludge. In addition, sludge-food waste AcoD regulates the C/N ratio and improves process stability for both substrates, as indicated by a low VFA/alkalinity ratio. Moreover anaerobic digestion of fruit and vegetable wastes can be performed without the addition of chemical alkali [46,189]. Co-digestion of sludge and food waste became effective at a ratio of 1:1 of volatile matter [190]. Methane yield increased from 20% to 40% in comparison with sludge monodigestion, with a loading rate reaching 3.83 kgVS.m^{-3}.d^{-1}. However, at higher OLR values, inhibition by affects digester performance. In another study, the addition of food waste to sludge at a ratio of 70% of TS improved methane production to 0.68 m^3.kgVS^{-1} in comparison with 0.42 m^3.kgVS^{-1} from sludge alone [177]. The total energy produced by sludge in the WWTP was estimated to 23 000 m^3 of methane per day, thus covering 42% of the energy demand of the plant. However, after co-digestion with food waste, a volume of 89 000 m^3 of methane was produced per day, almost 2-fold the energy consumed in the WWTP. Furthermore, Di Maria et al. [178] estimated the amount of recoverable energy from co-digestion of mixed sludge with fruit and vegetable waste in an existing sludge digester. The maximal methane production obtained after co-digestion was 900 NL. m^{-3} d^{-1} at an OLR of 2.1 kgVS m^{-3}d^{-1} and an HRT of 11 days. In these conditions, the electrical energy generated was 3500 MWh/year. In addition, the net costs of the plant were reduced by 37% after co-digestion of fruit and vegetable waste and sludge at a ratio of 60:40 [185]. Iacovidou et al.

Table 5
Properties and AD of sludge co-substrates reported in literature.

Substrate	Carbohydrates		Proteins		Lipids		Theoretical BMP (mL/gTS)	Experimental methane yield (mL/gTS)	Ref
	%TS	Contribution to BMP (mL/gTS)	%TS	Contribution to BMP (mL/gTS)	%TS	Contribution to BMP (mL/gTS))			
Theoretical methane potential[a]	100	415	100	496	100	1014	–	–	[161]
Primary sludge	55	228	18	89	10	101	418	213	[4]
Waste activated sludge	20	83	36	179	10	101	363	186	[4]
Food waste	62	258	19	94	14	142	494	510	[162]
OFMSW[b]	30–54	125–224	7–26	35–129	5–30	51–304	211–657	170–557	[163]
Fatty wastes	9	37	30	149	43	436	622	580	[164]
Dairy manure	35	144	17	84	0	0	228	51	[165]
Corn stover	72	299	5	25	0	0	324	241	
Wheat straw	69	288	2.5	12	0	0	300	245	
Rice straw	66	273	5.6	28	0	0	301	281	
Microalgae *Chlorella vulgaris*	21	87	16	79	41	416	582	420	[166]

[a] Theoretical methane potential of carbohydrates, proteins, lipids.
[b] Organic fraction of municipal solid wastes.

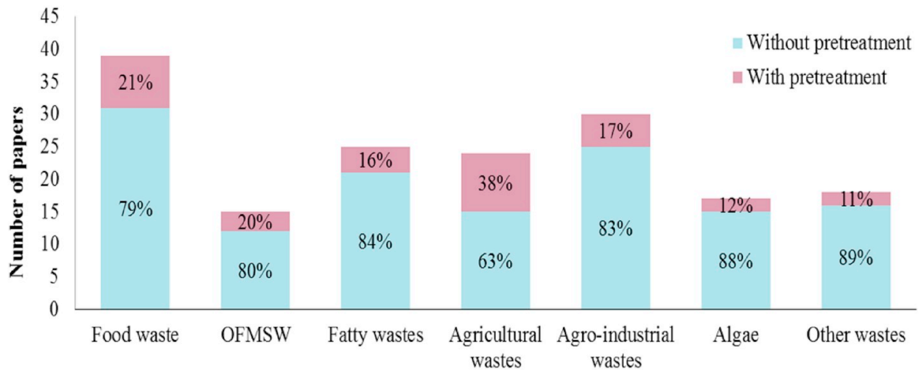

Fig. 3. Number of papers between 2013 and 2019 involving sludge (title) and anaerobic digestion (title) and methane (topic) (Web of science bibliometric study). Co-substrates were sorted manually.

[191] investigated the co-digestion of activated sludge mixed first with 10% of wasted cucumbers and secondly after the 127th day, with 10% of tomato waste. The methane yield rose by 75.7% compared to mono-digestion of the waste activated sludge. Besides the high methane potential of vegetable waste, this performance can result from the regulation of the C to N ratio, the dilution of heavy metals, and the presence of nutrients, such as phosphorus, which are essential for the activity of microorganisms and may compensate for the lack of nutrients in the co-substrate. However, co-digestion with food waste can also be limited by variability in the food waste composition which could disturb the process [191].

5.2. Fatty wastes

Fatty waste is a significant substrate for sludge AcoD, due to the high methane potential of lipids, although it can become inhibiting when the digester is overloaded. Luostarinen et al. [192] demonstrated that a semi-continuous sludge AcoD process is efficient when the grease trap sludge originating from a meat-processing plant is limited to 46% of volatile solids, for an HRT of 16 days and a maximum OLR of 3.46 kg VS. $m^3 d^{-1}$. In these conditions, the best methane yield was 463 m^3 t $VS_{added}{}^{-1}$ compared to 263 m^3 t $VS_{added}{}^{-1}$ from monodigestion of sewage sludge. Moreover, the addition of more than 60% (VS) greasy sludge entailed an accumulation of long-chain fatty acids (palmitic, stearic, and oleic acids) and subsequent process inhibition [168], a result which is in agreement with the findings of Luste and Luostarinen [171]. Li et al. [193] investigated the co-digestion of activated sludge with kitchen waste and FOG; it was found that FOG was an optimal co-substrate due to its high methane potential. Indeed, an increase from 123 mL .g VS^{-1} to 418 mL.g VS^{-1} was observed after 46% of FOG was added. However, lipids and fats can also hinder the mixture degradation: Martinez et al. [181] found that adding 0.2% of FOG to a semi-continuous reactor operating with 0.77 kg VS.$m^{-3}d^{-1}$, with a HRT of 30 days at 34 °C, decreased the methane yield by 22% in comparison with sludge alone. Using FTIR spectra, this result could be explained by the adsorption of lipids to the sludge cell walls.

5.3. Agricultural waste

Agricultural and lignocellulosic residues are also suitable for sludge co-digestion [194]. Olive pomace, wheat straw, rice straw and grass are examples of lignocellulosic co-substrates reported in literature. A mixture of olive pomace and sludge was co-digested at a ratio of 1:1 of VS and yielded 210 mL. gVS^{-1} while the methane produced in mono-digesters was 160 and 180 mL. gVS^{-1} for sludge and olive pomace respectively [179]. Wheat straw and activated sludge co-digestion was also reported in the literature. The methane produced was 26% higher than the sum of methane yields from both sludge and wheat straw at a ratio of 1:1 of VS [195]. Mixing rice straw and sludge was also synergetic, at a ratio of 1:1, the biogas yield obtained was 300 mL.gVS^{-1} while 225 and 188 mL.gVS^{-1} were obtained from monodigesters of rice straw and thickened activated sludge, respectively [196]. This synergy can be due to the regulation of the C to N ratio as well as the presence of nutrients in sludge which may fill the lack in lignocellulosic biomass. In addition, shredded grass from green spaces was subjected to co-digestion with sludge and higher biogas production was observed when 35% of grass was added, thus leading to a 36% enhancement in the methane yield compared to sludge mono-digestion [182].

5.4. Algal biomass

Microalgae and macroalgae species have also been co-digested with sludge. Costa et al. [175] investigated the co-digestion of two macro-algal species *Ulva* spp. *and Gracilaria* spp. No synergetic effect was observed on the methane yield. However maximal methane was obtained with *Ulva* added to sludge with a ratio of 15% TS [175]. In this case, the results from the co-digestion of sludge with the algae are neither positive nor negative. This may be explained by the features shared in common by microalgae and sludge. Both originate from wastewater treatment, while intracellular organic matter is hardly accessible. Co-digestion should reduce investment costs particularly. Instead of having 2 digesters, only one digester could be designed.

5.5. Agro-industrial wastes

Agro-industrial wastes such as milk and coffee waste [197], sugar beet pulp [198]and glycerol [199] have been used as co-substrates in previous works. Among them, glycerol has been most studied [183]. For a dose of 1% (v/v) of glycerol which is equivalent to a concentration of 385 µL.gVS^{-1}, the methane production increased from 354 mL.gVS^{-1} to 574 mL.gVS^{-1} of sludge. However, greater concentrations of glycerol can unbalance the digestion process. It is also noteworthy that the application of an organic load greater than 5.0 g COD.L^{-1}.d^{-1} has been observed to cause inhibition when glycerol is digested alone [200]. In another study, the addition of 2% (v/v) which corresponds to 590 µL.gVS^{-1} led to a 50% improvement of the methane yield at 37 °C [184]. Nevertheless, this improvement was only due to glycerol AD and not to a change in sludge degradability [201].

6. Discussion

Co-digestion of sludge offers environmental and economic advantages. Recent research on this topic has been reviewed in the present paper, showing that various types of organic waste can be co-digested with sludge. The main limitations of sludge anaerobic mono-digestion include i) their low methane potential, in particular in the case of secondary sludge, ii) their low C/N ratio and iii) in some cases, the

Table 6
Anaerobic co-digestion of sludge with various co-substrates in literature.

Substrates	Operating conditions	Methane from monoAD (mL/gVSadded)	Methane after coAD (mL/gVSadded)	Synergy (%)	Ref
Sewage sludge and sterilized solid slaughterhouse waste	Co-substrate (VS basis) 66% T (°C) 37 OLR (kg VS/m^3.d) 2.7 HRT(d) 20	Sewage sludge: 234 Sterilized solid slaughterhouse waste:719	619	+12	[167]
	Co-substrate (VS basis) 76% T (°C) 37 OLR (kg VS/m^3.d) 3.6 HRT (d) 22.5		585	−3	
WAS and greasy sludge from flotation process	Co-substrate (VS basis) 52% T (°C) 37 OLR (kg VS/m^3.d) 1.2 HRT (d) 24	WAS:269 Greasy sludge:917	574	−5	[168]
	Co-substrate (VS basis) 87% T (°C) 37 OLR (kg VS/m^3.d) 0.8 HRT (d) 25		166	−80	
PS and *Chlorella vulgaris*	Co-substrate (VS basis) 52% T (°C) 37 Batch	Primary sludge:531 Sewage sludge:160.6 *Chlorella vulgaris*:216	463	+26	[169]
WAS and *Chlorella vulgaris*	Co-substrate(VS basis) 90% T (°C) 37 Batch		204	−3	
WAS and *Chlorella sorokiniana*	Co-substrate(VS basis) 95% T (°C) 37 Batch	WAS: 362.3 *Chlorella sorokiniana* 318	442	+30	[170]
Sewage sludge and grease trap sludge from meat processing plant	Co-substrate(VS basis) 43% T (°C) 35 OLR (kg VS/m^3.d) 2.4 HRT (d) 20	Sewage sludge:300 Meat processing by-products:900	400	−28	[171]
	Co-substrate(VS basis) 64% T (°C) 35 OLR (kg VS/m^3.d) 2.9 HRT (d) 20		410	−40	
Sewage sludge and food waste	Co-substrate (VS basis) 53% T(°C) 35 OLR (kg VS/m^3.d) 7.2 SRT (d) 20	Sewage sludge:193 Food waste:439	332	+3	[172]
Sewage sludge and food waste	Co-substrate (VS basis) 58% T(°C) 35 Batch (d) 50	Sewage sludge:161 Food waste:400	367	+22	[173]
	Co-substrate (VS basis) 58% T(°C) 55 Batch (d) 50	Sewage sludge:157 Food waste:425	383	+23	
Mixed sludge and used oil	Co-substrate(VS basis) 28% T(°C) 37 OLR (kg VS/m^3.d) 0.91	Mixed sludge:342 Used oil:788	490.7	+5	[174]
Mixed sludge and macroalgae (*Ulva* spp.)	Co-substrate (VS basis) 11% T(°C) 37 Batch	Mixed sludge:335 Macroalgae:196	296	−7	[175]
Mixed sludge and grease trap sludge/OFMSW	Grease sludge (VS basis) 30% OFMSW (VS basis) 30% T(°C) 37 OLR (kg VS/m^3.d) 0.8 HRT (d) 20	Mixed sludge:300	547	–	[176]
Mixed sludge and grease trap sludge	Co-substrate (VS basis) 30% T(°C) 37 OLR (kg VS/m^3.d) 0.8 HRT (d) 20	Mixed sludge:300	456	–	
Sewage sludge and food waste	Co-substrate (VS basis) 71% T(°C) 35 OLR (kg VS/m^3.d) 1.77 HRT (d) 30	Sewage sludge:288	462	–	[177]
Sewage sludge and fruit and vegetable wastes	Co-substrate (VS basis) 59% T(°C) 35 OLR (kg VS/m^3.d) 2.8 HRT (d) 10	Sewage sludge:102	267	–	[178]
Sewage sludge and olive mill waste	Co-substrate (VS basis) 40% T(°C) 37 Batch (d) 30	Sewage sludge:160 Olive waste:180	210	+19	[179]
Sewage sludge and cow manure	Co-substrate (VS basis) 46% T(°C) 35 Batch (d) 63	Sewage sludge:251 Manure:319	328	+16	[180]

(*continued on next page*)

Table 6 (*continued*)

Substrates	Operating conditions	Methane from monoAD (mL/gVSadded)	Methane after coAD (mL/gVSadded)	Synergy (%)	Ref
Sewage sludge and fats, oils and grease	Co-substrate (VS basis) 6.5% T(°C) 35 OLR (kg VS/m^3.d) 0.77 HRT (d) 30	Sewage sludge:304	298	–	[181]
Sewage sludge and shredded grass	Co-substrate (VS basis) 35% T(°C) 35 Batch (d) 14	Sewage sludge:493	432	–	[182]
Sewage sludge and glycerol	Co-substrate (VS basis) 0.4% T(°C) 35 OLR(kg VS/m3.d) 0.78 HRT(d) 24	Sewage sludge:354	574	–	[183]
Sewage sludge and glycerol	Co-substrate (VS basis) 2% T(°C) 37 OLR(kg VS/m3.d) 3.68 HRT(d) 17	Sewage sludge:425 Glycerol:483	450	+6	[184]
Sewage sludge and fruit-juice industrial waste	Co-substrate (VS basis) 38% T(°C) 35 Batch(d) 20	Sewage sludge:438 Fruit-juice waste:382	306	−26	[185]

presence of heavy metals. Using co-substrates with a higher methane potential than sludge should systematically increase the overall methane yield of the plant. This applies for food waste, fatty waste and agro-industrial waste (in particular glycerol). The low C/N ratio of municipal sludge digestion alone can lead to system instability. Co-digestion with low nitrogen content residues, such as lignocellulosic residues, agro industrial residues or certain types of fatty waste improves the nutrient balance in the whole reaction process, which is conducive to the stability of the system and can induce synergy between both substrates. Finally, when the heavy metal content in sludge is high and may induce the inhibition of anaerobic digestion, co-digestion with any type of waste that does not contain heavy metals will be beneficial as it would reduce the heavy metal concentration by simple dilution.

Considering co-substrate, the benefits of co-digestion with sludge are intake of water, micronutrients and buffer capacity by sludge. These are particularly useful for lignocellulosic residues, municipal solid waste as well as certain kinds of food waste. Also, another advantage of co-digestion with sludge is the adjustment by a low C/N ratio, which is particularly important for lignocellulosic residues, paper and cardboard wastes and certain agro-industrial and fatty wastes.

Finally, it is noteworthy that co-digestion efficiency is strongly related to feedstock availability and variability, transportation costs, digestate effective disposal and legislative restrictions. In particular, the use of sludge as a co-substrate implies harsher regulations for digestate land application.

Co-digestion of OFMSW and food waste has been most studied and most applied at full scale [202,203]. To enhance methane production, lipids and glycerol can also be useful although their ratio in the feedstock should not be too high. When the mono-digester already exists in the WWTP, the only extra costs of co-digestion are those of co-substrate transport. Thus, by optimizing the OLR and mixture ratio, co-digestion can allow for the entire consumed energy in the WWTP to be retrieved. Co-digestion performance can also be enhanced with feedstock pretreatment.

7. Combination of pretreatments and sludge co-digestion

Co-digestion benefits process stability by adjusting the C/N ratio, moisture content and by diluting inhibitors. However, to improve AcoD effectiveness, pretreatments can be applied to all substrates, either separately or mixed. Fig. 3 presents the number of peer-reviewed papers investigating the effects of pretreatment on the AcoD of sludge and different organic wastes used. It is obvious that the combination of AcoD with pretreatment has been scarcely studied when compared with AcoD without pretreatment; the main reason for this might be related to the additional costs it implies. Pretreatments combined with co-digestion are reported in Table 7.

7.1. OFMSW and food wastes

Following an alkaline pretreatment of OFMSW with sodium hydroxide at a dose of 6% TS and its co-digestion with 40% sludge, the methane production rose by 20% ($p < 0.05$). VS reduction was also enhanced and reached 67% compared to 50% for the untreated mixture ($p < 0.01$) [211]. Besides chemical pretreatment, fungal mash was used to pretreat a mixture of sludge and food waste. This resulted in a higher methane production compared to another approach where sludge was mixed with the liquid fraction of food waste pretreated with fungal mash [209]. In addition, biological pretreatment was performed on the mixture of sludge and food waste with the addition of inoculum in the absence of oxygen at 35 °C, and for different pretreatment times. After 24 h of pretreatment (the optimum duration), the hydrolysis rate increased by 20%, soluble COD and soluble polysaccharide concentrations increased by 130% and 60% respectively compared to the untreated mixture, while the methane yield was 25% higher [210]. This increase in hydrolysis rate implies that the pretreatment had a higher impact on sludge, as food waste is easily hydrolysed. Another study compared thermo-alkali, thermal, alkali and ultrasonic pretreatments applied to food waste, waste activated sludge and their mixture composed of 70% FW and 30% sludge. Ultrasonic and thermal pretreatments produced higher methane yields (+69% and +65% respectively) and a stronger reduction in VS when applied to the mixture [213]. When food waste was pretreated alone before sludge addition, a mechanical pretreatment followed by thermal pretreatment was found to be the optimal method for solubilising a greater amount of sugars [214]. When the mixture was pretreated, thermo-alkaline pretreatment led to a potentially higher solubilisation rate of sugars, proteins and lipids, to VS reduction and to a higher methane yield. Zhang et al. [204] reported that a mixture of sludge subjected to microwave pretreatment led to a better methane yield than a mixture with microwave-pretreated food waste. This was related to the accumulation of propionic acid in the latter mixture. Sludge was also subjected to alkaline pretreatment prior to its co-digestion with food waste: the addition of 45 mEq of NaOH per litre increased methane yields by 66%, 73% and 88% at 25 °C, 35 °C and 55 °C [215].

Table 7
AcoD of sludge combined with pretreatments in literature.
References cited: [169,179, 196, 204–212].

Substrates	Pretreatment of sludge	Pretreatment to cosubstrate	Conditions	Methane after AcoD(mL/gVS$_{added}$)	Methane after AcoD+ pretreatment (mL/gVS$_{added}$)	Ref
Sewage sludge and food waste (1:1)	Microwaves		600W at 100°C (2,45 MHz), Batch AD (33days)	297	318	[204]
		Microwaves			311	
WAS and olive waste (1:1)	Microwaves		Microwaves (175°C,30min,2MPa), Batch AD at 37 °C	210	320	[179]
	Ultrasonic		US (200W,15min,20kHz), Batch AD at 37 °C		260	
			US (200W,30min,20kHz), Batch AD at 37 °C		260	
WAS and fatty wastewater (33/67)	Thermoalkaline		120°C, pH=8 ; 9 ; 10 Batch AD	542	565 577 582	[205]
			80°C, pH=8 ; 9 ;10 Batch AD		576 for all pH values	
WAS and fatty wastewater (90/10)			80°C, pH=8, Semi-continuous AD	172	271	
WAS and fats, oil and grease (1:1.2 TS)	Thermoalkaline		T=55°C and pH=10, modified 2 stage thermophilic digester(55°C)	262	288	[206]
Mixed sludge (30% PS and 70% SS) and glycerol (1% v/v)	Alkaline		pH=12 Continuous AD at 35°C	500	70	[207]
WAS and tobacco wastes (C/N of the mixture 24/1)	Ozone		90 mg/gTSS Batch AD at 35°c	204	281	[208]
WAS and food waste (1:1)	Biological		1.5 g of dry weight of fungal mash per liter; at 60°C for 8h BMP at 35°C	nd	743	[209]
	Biological		Mixture seeded with inoculum for 24h at 35°C in the absence of oxygen Batch AD at 35°C	230	294	[210]
WAS and OFMSW (40:60)		Alkaline	NaOH ,6% for 24h at 25°C BMP at 37°C for 18 days5% of TS,	263	337	[211]
Thickened WAS and OFSMW and rice straw (0.5:3:0.5)	Thermal		70°C for 9h Batch AD at 37°C for 55 days.	TWAS 190 OFSMW 214 RS 230	530	[196]
		Alkaline	NaOH: 3% (w/w) Batch AD at 37°C for 55 days.		538	
	Thermoalkaline	H_2O_2	For sludge: NaOH, pH=11 for 10h and 90°C For Rice straw: H_2O_2; 3% (w/w) Batch AD at 37°C for 55 days.		558	
PS and *Chlorella vulgaris* (3:1)		Thermal	16g/L of microalgae at 120°C for 40 min. Batch AD at 35°C for 30 days.	462	509	[169]
Sewage sludge and oily wastewater (40 :60)		Hydrodynamic cavitation	40Wh/L Batch AD at 35°C for 30 days.	687	894	[212]

7.2. Fatty wastes

Fat, oil and grease were pretreated at pH = 10 and 55 °C and then mixed with sludge. Methane production rose by 34%. This pretreatment increased soluble COD from 50% to 76% [206]. A mixture of fatty wastewater and sludge was pretreated within a range of pH between 8 and 10, and under three temperature conditions: 80 °C, 120 °C and 170 °C. Thereafter, pretreatment at pH = 8 and 80 °C on a mixture of 90% sludge and 10% fatty wastewater increased the methane yield by 58% [205]. It appeared evident that soft pretreatments at mild temperatures were most suitable for fatty wastes and their mixtures with sludge. Li et al. [212] reported a 10% increase in methane production following thermochemical pretreatment of a mixture of sludge and fats, oils and grease (Table 7). Hydrodynamic cavitation was also employed to pretreat oily wastewater. The mixture of 40% sludge with 60% pretreated oily wastewater increased the methane yield from 687 mL/gVS to 894 mL/gVS.

7.3. Agricultural wastes

Rice straw and OFMSW were added to sludge for co-digestion. Different pretreatment conditions were applied to rice straw and sludge. The optimal scenario was the thermo-alkaline pretreatment of sludge and H_2O_2 addition to rice straw, which yielded 558 mL/gVS with a VS removal of 77% [196].

7.4. Algal biomass

It can be efficient to improve cell and lignin disintegration of algae by first reducing the particle size before its co-digestion with sludge. In Tedesco et al. [216], algal biomass (*P. Caniculata*) was mechanically pretreated by a Hollander beater (for 10 min), then codigested with sludge. The resulting methane production was 20% higher than the control with an 85% gain of energy. Concerning microalgae in particular, Mahdy et al. [169] observed that a mixture of 25% thermally pretreated secondary sludge and 75% microalgal biomass, produced the highest methane volume, although this remained lower than their AcoD excluding pretreatment.

8. Discussion

Co-digestion combined to pretreatment can significantly improve methane production if the pretreatment is correctly selected. Pretreatment should only be carried out on substrate that is most difficult to degrade. Indeed, the pretreatment of an entire mixture would result in additional costs (chemicals and energy consumed). In particular, food waste and glycerol which present a high methane potential and hydrolysis rate do not require pretreatment. Furthermore pretreatment of glycerol [201] or mixing with pre-treated sludge could even lead to inhibition due to the too high amount of very accessible matter (Table 7). However, energy recovery from OFMSW does improve after biological or thermal pretreatments (< 100 °C) [217]. Indeed, lignocellulose-rich agricultural waste become more easily hydrolysed and digested when its accessibility is improved (e.g. grinding and extrusion) and when it is delignified (e.g. alkaline pretreatment and ozonolysis), while hemicelluloses and cellulose can be degraded through biological pretreatment [218]. Likewise, thermoalkaline pretreatment is widely employed for fatty waste as it favours their

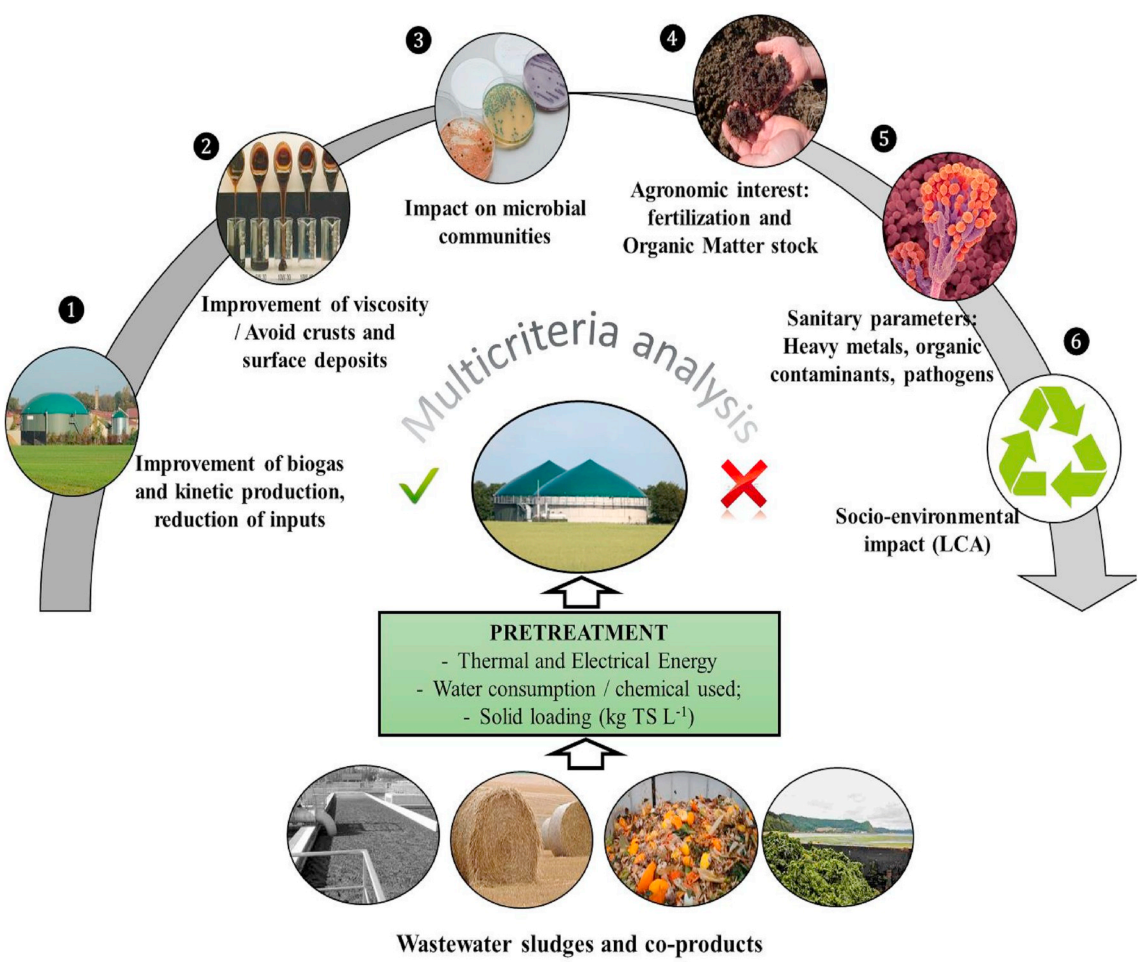

Fig. 4. Schematic of the proposed multi-criteria analysis aiming at assessing the potential benefits and the overall sustainability of the pretreatment steps.

solubilisation by saponification and thus their contact with microorganisms. It can enhance AD stability by preventing LCFA accumulation. Microalgae can be subjected to mechanical or low temperature pretreatment to improve their methane production. It is important to mention here again that the selection of conditions is strongly related to the type of sludge and to its co-substrate properties, to costs and energy consumption considerations as well as the digestate disposal strategy.

9. Research outlooks

9.1. Perspectives

Anaerobic digestion stands out as one of the most promising options for wastewater sludge management and valorisation, as it improves the overall sustainability chain. It is a mature and generally cost-effective technology, providing energy production and efficient disposal of sludge with possible nutrient recovery from the digestate. Energy efficiency can be improved by co-digestion with various organic residues. Furthermore, pretreatment seems to be a promising route for improving the accessibility and biodegradability of wastewater sludge and co-products applied in an anaerobic co-digestion process. Nonetheless, until present, pretreatment efficiency has generally only been taken into account from an energy saving and economic point of view [178–180].

For this purpose, energetic and economic indicators have been developed to evaluate whether the excess biogas produced can cover or not the energetic and economic requirements of pretreatment [220,221]. Several factors can affect the overall balance of pretreatment, such as biogas valorisation types (CHP or injection), national incentive policies for electricity or biomethane injection, operational parameters of a given pretreatment (e.g. solid loading, temperature, chemical reagent), energy and heat integration at the scale of a wastewater treatment plant.

It remains insufficient to assess the sustainability of applying a pretreatment, if only considering the gain in energy compared to a conventional baseline (AD without pretreatment). Fig. 4 exhibits the different possibilities of the proposed multi-criteria analysis scheme that can be potentially applied to assess the true benefits of a pretreatment step prior to an AD process. With this type of multi-criteria analysis, the benefits of a pretreatment step prior to AD process should be assessed according to various technical, microbial, agronomic or environmental aspects. Several aspects can be considered such as:

i) the energetic/economic benefits, which are usually addressed;
ii) an improvement of viscosity. Pretreatment technologies can modify the rheology/viscosity of the biomass mixtures, reducing energy requirements for pumping and mixing. They can also contribute to the reduction of crusts and surface deposits. Several pretreatment technologies such as mechanical, thermal and enzymatic treatments have displayed their advantages for improving rheological properties [222,223]. For instance, hydrodynamic cavitation was reported to influence viscosity, thus contributing to reducing the energy demand for mixing, heating and pumping [222]. Similarly, enzymatic pretreatment has been recognized to lower the viscosity of substrates [219]. In parallel, pretreatment has also been shown to contribute to the improvement of the dewaterability of digestate following the anaerobic digestion process [224];
iii) the impact on microbial communities and performances: anaerobic digestion (AD) performance wholly depends on the activity of microbial communities. A better knowledge of anaerobic microbial communities could promote the development of efficient microbial consortia for enhancing AD processes [225,226]. Pretreatment technologies could impact microbial communities in either a negative or positive way. So far few studies have genuinely investigated the impact of pretreatment on microbial communities [225,226] and it should certainly become a major R & D axis in the coming years. The impact of pre-treatment on performance/stability of the AD process should thus be better understood;
iv) agronomic interest: fertilisation and stock of organic matter. Pretreatment technologies can also affect the nutrient content and the organic matter stability of the digestate [227]. At present, few studies have investigated the impact of pretreatment technologies on such parameters. Some useful tests such as carbon and nitrogen mineralization but also germination and plant growth tests can be proposed to evaluate these parameters [29,227];
v) sanitary parameters: for agronomic issues, pretreatment technologies can positively or negatively affect the sanitary impacts of digestate. Indeed, thermochemical pretreatment can contribute to increase the content directly (by chemical addition, Na, S, Fe) or indirectly (through formation of by-products such as furans and polyphenols) of certain elements in the digestate that might cause detrimental impacts on the soil [228,229]. On the contrary, pretreatment can also affect the pathogen content, the antibiotic resistance and lower the amount of organic contaminants [227,230]. Recently, Tigini et al. [231] defined a protocol applying seven ecotoxicity tests and considering ten different endpoints in order to evaluate its impact on the environment and human health. It is therefore strongly recommended to perform a detailed analysis of these various sanitary parameters on the digestate of pretreated feedstock;
vi) the environmental impact: Loustau-Cazalet et al. [232] evaluated the amount of water, and chemical reagent used during biomass pretreatment. A specific factor called 'En' was developed, defined as the ratio of the mass of waste per unit of product. Although such factors can allow for the quantity of waste to be evaluated in comparison with generated products, they cannot allow for a general impact on environmental items to be assessed, as is possible with Life Cycle Analysis (LCA). LCA is a technique for assessing environmental impacts associated with each stage of the valorisation chain. Presently, few studies have yet investigated the environmental impact of pretreatment prior to the AD process [196–198]. Different pretreatments applied to two types of waste (kitchen waste and sewage sludge) have been environmentally evaluated using a life cycle assessment (LCA) methodology [233–235].

In general rules, anaerobic digestion of wastewater sludge in co-digestion appears to be a promising solution for waste management. Nonetheless, during the AD process, part of the matter does not degrade and remains in the digestate [236,237]. Digestate is essentially composed of water (more than 90%), nutrients, residual undegraded fibres (e.g. lignin, cellulose), and inorganic compounds (e.g. ash) [236,237]. Due to the low HRTs (Hydraulic Retention Times) that are generally applied in industrial biogas plants, after which biogas production starts to decrease, part of the organic matter still remains within the solid phase of the digestate [238,239]. Digestate can generally be separated into a liquid and a solid phase that are then used for their fertilizing or amending properties [236,237]. However, the use of digestate in agronomy is not always suitably adapted to the geographical context, especially in intensive farming areas. A number of shortcomings have recently been reported, including potential risks of water pollution through leaching, soil contamination, or even a threat to human health by food contamination [237,240]. For this reason, several alternative valorisation routes for both liquid and solid digestates have been investigated during the past decades, in order to improve the environmental sustainability of the overall process [236,237]. As illustrated in Fig. 5, interest is focused on the nutrient-rich liquid digestate for use in the cultivation of microalgae [241,242]. In turn, microalgae could be further valorised for biodiesel production [243,244], returned to the AD process with or without prior biodiesel extraction [243,245] or used as slow release fertilizers [246,247].

Concomitantly, the fibre-rich solid fraction could potentially be used in biorefinery processes [239]. Composting has also been regarded has a feasible treatment for stabilising digestate and thus improving its properties as a soil conditioner or substrate [248,249]. Recently, valorisation of solid fraction of digestate through a pyrolysis process, gasification and combustion has been reported. Indeed, pyrolysis and gasification have been widely investigated these past few years with several synergies and complementarities between the two processes that have been reported [236,250–252].

It is obvious that thermochemical processes (i.e. pyrolysis, gasification, combustion) can represent promising alternative valorisation routes even though they still require a drying step of the solid digestate before processing. Most European agricultural biogas plants are currently converting biogas in a Cogeneration Heat and Power (CHP) system. Generally, a large proportion of heat is lost due to the remoteness of farms that prevents its distribution to the district. Recently, the excess heat produced during anaerobic digestion has been shown to cover the drying needs for solid digestate [91]. However, the development of units with biomethane injection might limit the valorisation of solid digestate through thermochemical processes, as it does not produce any excess heat. The social aspect should also not be neglected. It is imperative for citizens, and especially farmers, to be fully aware (to avoid the "not in my backyard" (NIMBY) syndrome) of the importance of implementing digesters and ancillary units for pre-treating feedstock and post-treatment digestate. Their involvement is crucial in the use of biogas energy and water for irrigation, for example, and should be ensured. Legislation is also an essential aspect, as it can regulate the authorisations for setting up AD units and contribute to adequately manage their waste and gaseous emissions [253].

9.2. Practical implications of this study

AD represents one of the most mature existing processes that are capable of reducing sludge volumes and has the capacity to generate biogas. The co-digestion of sludge with other organic wastes has the advantage of improving methane production by adjusting the C to N ratio for example. However, co-digestion is presently facing challenges such as feedstock variability, transportation costs, and legislation guidelines. Generally, sludge treatment using an AD process represents a promising pathway that should be integrated into a biorefinery concept for the overall system to become more sustainable. This can be ensured thanks to: i) a progressive integration ("industrial symbiosis") of different conversion technologies, establishing functional connections and links across different processes and ii) the implementation of a "cascading" biomass utilisation scheme, where the output of one process becomes the input of the following one (thus also targeting the "zero-waste" goal), with biomass progressing through a series of material flows and conversions. The main objectives of such an approach should be to comply with several sustainable pillars set up by European policies and the 2015 Circular Economy Action plans. These involve maximizing the reuse of waste, creating value from waste and developing new business models and jobs. European legislations propose environmental guidelines for the sustainable valorisation of organic wastes in EU countries and these evolve with time. In contrast, developing countries lack regulations for governing the management of waste, and especially sludge. Indeed, the establishment of laws limiting landfilling and improper sludge dumping should force manufacturers to seek clean and sustainable methods for disposing of their waste. In turn, this should encourage advances in scientific research. Finally, the influence on policy makers is not only driven by the economic context but also by environmental and societal purposes. As mentioned by Manara and Zabaniotou [254], sustainability is held by three pillars: they are economic, environmental and social. Future implementation of the concepts defined in this review does not solely depend on technical constraints: a multidisciplinary approach integrating technical, social, environmental and economic sciences still needs to be implemented. Furthermore, the coming territorial monetarisation of social and environmental benefits of novel approaches should allow for a monetarised gain of the impacts as well as an influence on stakeholders and policy makers involved in waste treatment and valorisation.

10. Conclusion

Biological and thermochemical processes besides other novel advanced processes can be applied to valorise sludge. Conversely, sludge

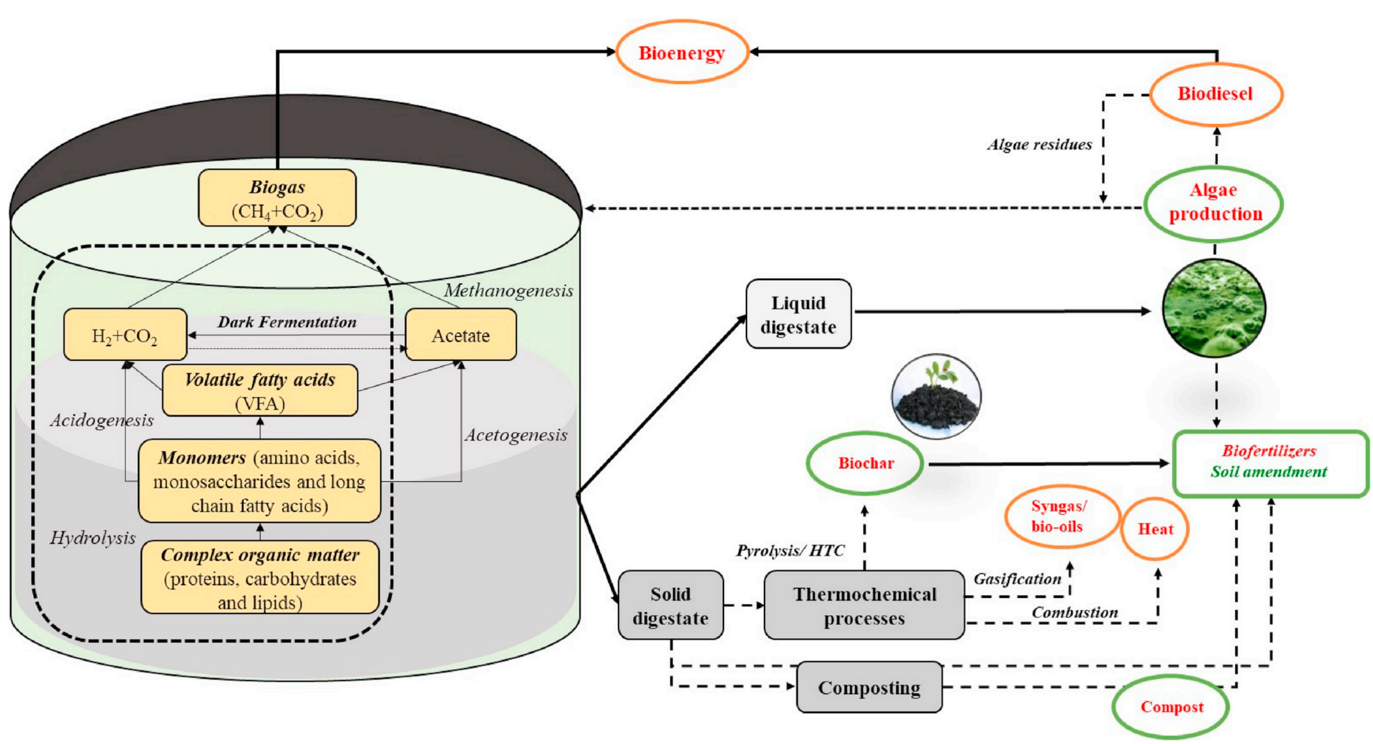

Fig. 5. Integration of the classical wastewater AD process into a biorefinery scheme.

represents a low-cost material for these processes. In particular, sludge co-digestion aims at enhancing methane production by adjusting several factors such as pH, moisture, the C/N ratio and nutrient availability. The efficiency of AcoD and pretreatment combination is not guaranteed. Indeed, it necessarily depends on the pretreatment conditions and on the nature of the co-substrates (e.g. lignocellulosic or fatty waste). In general, further studies focusing on the synergetic effects of combining pretreatment and AcoD processes are still required for a better understanding of the interactions between substrate properties and their impacts on the microbial communities and digestate properties.

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