

Review

High-Solid Anaerobic Digestion: Reviewing Strategies for Increasing Reactor Performance

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Abstract: High-solid and solid-state anaerobic digestion are technologies capable of achieving high reactor productivity. The high organic load admissible for this type of configuration makes these technologies an ideal ally in the conversion of waste into bioenergy. However, there are still several factors associated with these technologies that result in low performance. The economic model based on a linear approach is unsustainable, and changes leading to the development of a low-carbon model with a high degree of circularity are necessary. Digestion technology may represent a key driver leading these changes but it is undeniable that the profitability of these plants needs to be increased. In the present review, the digestion process under high-solid-content configurations is analyzed and the different strategies for increasing reactor productivity that have been studied in recent years are described. Percolating reactor configurations and the use of low-cost adsorbents, nanoparticles and micro-aeration seem the most suitable approaches to increase volumetric production and reduce initial capital investment costs.



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1. Introduction

Anaerobic digestion is a technology that is widely applied for the treatment of biowastes. This process can degrade organic components in the absence of oxygen, generating biogas and the digested material as a residual stream (digestate). Digestate contains the remaining solids that are not susceptible to microbial degradation under anaerobic conditions, humic and fulvic substances, cell material, and nutrients. The stabilized organics derived from this process, show characteristics that depend on the input materials and the performance of the reactor. This liquid digestate can be considered agricultural wastewater, with interesting potential for the recovery of nutrients and humic substances [1]. Digested solids are also a valuable organic amendment, and their application to agronomic lands allows for the recycling of nutrients. Digestate can act as a soil improver and has therefore been confirmed as a valid resource for sustainable management [2,3].

Biogas is a valuable product that is also generated from the anaerobic decomposition of organics, containing methane and carbon dioxide as major components. When evaluating the digestion performance, the amount of biogas produced and the removal of organic materials are relevant parameters. The outcome of the process is highly dependent on the organic loading applied to the reactor, and this loading in turn determines the treatment capacity of the system. The quality and organic concentration of the feed impact the economic feasibility of the digestion process as they have a direct effect on daily biogas production and the volume of the reactor. One of the main obstacles impeding the wider implementation of this technology is high investment costs [4]. Therefore, it is crucial to

attain a significant increase in volumetric reactor production without negatively affecting digestate quality.

The valorization of biogas for the production of energy or to upgrade this gas to achieve a quality similar to that of natural gas is another integral component of digestion technology. Biogas conversion or upgrading also has a clear impact on the capital investment and operating costs of this technology. Energy production from biogas is usually performed by combined heat and power units (CHP), allowing the efficient use of on-site biogas [5]. The applications of fuel cells and micro-turbines are increasing, but the costs associated with these later technologies are still too high.

A large amount of water in wet digestion systems translates into lower methane productivity [6] since biogas yields are directly associated with the dry matter content of the feeding. Thus an increase in reactor productivity usually translates into increasing the solid content of the input material. Increasing the solid content in the digestion system exerts different effects on microbial performance; thus, the term “high-solid” is used to refer to an anaerobic process that are still considered wet digestion systems but which work with solid content values close to the higher limit of this solid range, and experience diffusion limitations [7,8]. High-solid anaerobic digestion seems to be a logical option for enhancing digestion performance, given that biogas production is directly associated with the mass of volatile solids fed into the digester. However, the strategy of working with a higher solid content implies a great variety of modifications in plant operation and the equipment needed, and the higher organic matter content significantly affects the performance of the anaerobic microflora.

Wet digestion, on the other hand, is easier to operate. The feeding material needs to be diluted with water to attain a desired solid content to prevent clogging or dense scum formation in the reactor liquor, which would otherwise lead to deficiencies in mixing and in biogas generation [9]. The increase in the feeding solid content has a marked impact on waste rheology and, therefore, on the equipment needed for transporting and mixing the liquor inside the reactor. The operating temperature of the digestion process also affects digestate rheology, and it was reported by Dai et al. [10] that thermophilic digestate presented better flowability than mesophilic digestate, probably because it has lower free and interstitial moisture. Residence time also affects sludge behavior; thus, longer digestion times aid in decreasing yield stress [11], favoring pumping characteristics.

The addition of water is necessary to set a specific solid content for the feeding, and it is usually recirculated to avoid excessive consumption of this resource. The liquid digestate may be treated for the recovery of nutrients through struvite precipitation [12,13] and solids can find applications as organic amendments in croplands. However, the amount of solid digestate produced is still high, and it may prove difficult to ensure a proper final disposal option all year round. Thus, alternatives are needed for increasing the conversion of the degradation process and the final valorization of the digestate to avoid generating an additional problem for farmers with no possibility of finding a solution for the final disposal of the digestate. Anaerobic reactors capable of treating highly concentrated substrates without experiencing significant inhibitory problems would aid in increasing plant feasibility, facilitating digestate handling operations, and final disposal.

Anaerobic digestion can be described as a sequential process in which complex materials are initially hydrolyzed and then transformed into short-chain molecules, in a series of intermediary stages, in which volatile fatty acids are produced along with other compounds such as hydrogen, alcohols, and formate [14,15]. This sequence has a great effect on the final reactor performance, and the balance of the different reactions involved in this sequence is extremely crucial for high-solid digestion systems. Methanogenic reactions are responsible for the production of biogas and this last stage must match perfectly in a syntrophic interaction [16], thus avoiding the accumulation of undesirable compounds.

Particulate substrates experience digestion limitations associated with the time needed for solubilizing the organic components to make them accessible to microorganisms. The particle size affects the time required to complete the first hydrolysis stage and this is

explained by physical restrictions associated with the specific surface area available to be attacked by enzymes [17]. For this reason, pre-treatments are usually employed to facilitate this stage, with thermal pre-treatment and ultrasonication being the predominant technologies used at a commercial scale. Several pre-treatment methods have been evaluated with success under laboratory conditions, achieving a significant enhancement in the solubilization of organics. However, the capacity of thermal pre-treatments to recover heat makes these superior when considering their performance in terms of the energy balance [18].

The traditional digestion process must be performed with careful control to avoid overloading and acidification problems, and this feature is even more relevant in high-solid digestion systems. The process is usually evaluated under optimal conditions at the laboratory scale, but these optimal conditions may not be at all feasible in large-scale operations. When high-solid digestion systems are studied under simplified configurations with low mixing, the treatment capacity is significantly reduced, leading to operation under low organic loadings [19] and high residence time. Digester heating is also necessary to maintain an adequate conversion level, but this creates an excessive economic burden regarding the installation and operating costs. Therefore, suitable heating technologies, capable of reducing digester energy demands, are also strongly recommended.

In the present manuscript, a description of the performance of anaerobic digestion is reviewed, considering the effect of increasing the solid content in the reactor to increase biogas productivity. This review aims to offer a description of the different alternatives studied regarding the use of supplements for reducing the toxicity associated with the accumulation of intermediary compounds and ways of ensuring high performance in high-solid operations. This manuscript describes the effect of increasing solid content in digestion systems along with the particularities of operating under solid-state configurations. It is also reviewed the different strategies evaluated in the literature for attenuating adverse effects associated with the accumulation of inhibitory compounds and the impact of temperature in digestion reactors.

2. The Effect of Organic Loading on Digestion Performance

High-solid anaerobic digestion (HS-AD) and solid-state anaerobic digestion (SS-AD) are technologies presenting an outstanding capacity for treating organic wastes and requiring lower digester volumes. The term “solid-state digestion” is usually used when the dry matter content of the feeding material exceeds 15% [20], whereas HS-AD can be defined based on the limit established by Zhang et al. [7,8] with regard to diffusion behavior, indicating that total solid content greater than 6% represents the boundary between low and high solid digestion.

However, the increase in solid content results in several negative effects, leading to imbalances in the process. Xu et al. [21] reviewed the performance of HS-AD of sewage sludge, also considering 6% total solid content as a barrier for defining HS-AD. They indicated that the main limitation when operating under these conditions was associated with process instability due to mass transfer limitation problems, high viscosity, and the accumulation of inhibitory compounds. The increase in solid content, and therefore the reduced water phase, causes the accumulation of volatile fatty acids (VFAs) and ammonia, decreasing the methane production rate [22]. Improvements in mixing may aid in reducing mass transfer limitations, but then the amount of energy needed to provide a suitable mixing rate in some cases may become excessively high, given the rheology of sludge.

In anaerobic reactors working as a completely stirred tank reactor (CSTR), cell retention time and hydraulic retention time (HRT) are coupled. The degradation of complex wastes may require a higher time inside the reactor to attain their full conversion, but on the contrary, simple organics are readily degraded. When digesters operate as a CSTR, the decrease in the residence time is also accompanied by an increase in the organic loading rate (OLR) due to the higher incoming flow. This effect may translate into higher biogas production thanks to the greater amount of fresh material loaded. However, the system

may experience preferential degradation of the substrate and incomplete degradation of the complex material, leading to lower gas yields and the incomplete stabilization of organics [23]. Considering that anaerobic digestion is a process intended to reduce the putrescible potential of organic materials and attain waste stabilization, any decrease in the organic quality of the digestate should be considered an undesirable feature.

Anaerobic digestion involves several reaction mechanisms, in which anaerobic bacteria and archaea species transform biomass into a sequence of biological reactions [24]; thus, any change in operating conditions directly affects the microbiology of the system and the final outcome of the process. Ziganshin et al. [25] and Langer et al. [26] studied the correlation between microbial communities and parameters determining reactor performance, indicating that temperature, ammonia content, and the type of substrate had a marked effect on the diversity of these communities, fungi, and archaea organisms.

The relationship between HRT, OLR, and ammonium content is complex, since inhibitory effects may be conditioned to the capacity of the microflora to degrade proteins during the time available inside the digester. Therefore, ammonia content, VFA evolution, and residence time are closely linked, based on the operating conditions. Table 1 shows the results from different authors reporting biogas yields at different OLRs applied under mesophilic regimens.

Table 1. Results reported in the literature on the digestion of different substrates under mesophilic conditions, varying the organic loading rate (OLR) and reporting the ammonia content in the reactor.

Substrate	OLR (g VS/Lr d)	Ammonia (TAN) (mg/L)	HRT (d)	Methane Yield (L/g VS)
Slaughterhouse waste and food wastes [27]	0.9	2143	50	0.53
	1.16	3022	36	0.64
	1.7	3210	25	0.56
	1.85	2106	50	0.4
	2.56	3830	36	0.45
	3.7	4099	25	0.5
High-solid digestion of sewage sludge [28] ¹	3.0	3250	30	0.27
	3.5	3176	25	0.24
	4.0	2635	20	0.18
	4.5	1968	17	0.18
	5.0	2585	10.5	0.18
	7.0	2596	6	0.15
	8.5	2255	4	0.12
3.0	3054	30	0.25	
Blood and food wastes [29]	1.5	1921	36	0.2 ²
Swine and poultry manure co-digestion with sewage sludge [30]	1.27	1066	30	0.21 ²
	1.91	1174	20	0.27 ²
	1.43	1189	30	0.20 ²
	2.15	1261	20	0.18 ²
	2.86	1264	15	0.23 ²

¹ Ammonia values were digitized from graphs reported in the reference. ² Value calculated from data provided using biogas production and methane content.

Due to the great variety of ways of describing reactor performance, the homogenization of values is not easy; thus, few references can be compiled in a single table. In the present case, the values shown in Table 1 were comparable in that they were obtained under semi-continuous conditions, with the authors reporting the organic loading rate (OLR) and the equivalent hydraulic residence time (HRT), in which ammonia and methane yield can be extracted. To better visualize the information reported in Table 1, the data were graphically represented using a ternary diagram (represented in Figure 1) and normalizing values (listed in Table 1) to unity. Extremes were added to facilitate visualization, assuming the maximum methane yield as the value reported in references when considering min-

imum OLR or maximum HRT. We also assumed a 50% inhibition when considering the maximum ammonia content, based on the results reported by Poggi-Varaldo et al. [31]. Several factors affect reactor performance and this graph should be carefully interpreted so as not to extrapolate erroneous conclusions. There exists a relationship between volatile solid content, nitrogen content, and operating conditions. This hypothesis was evaluated by Rattanapan et al. [32], indicating that the C/N ratio and OLR can be optimized based on reactor operating conditions, obtaining the maximum biogas yields for specific values of these parameters.

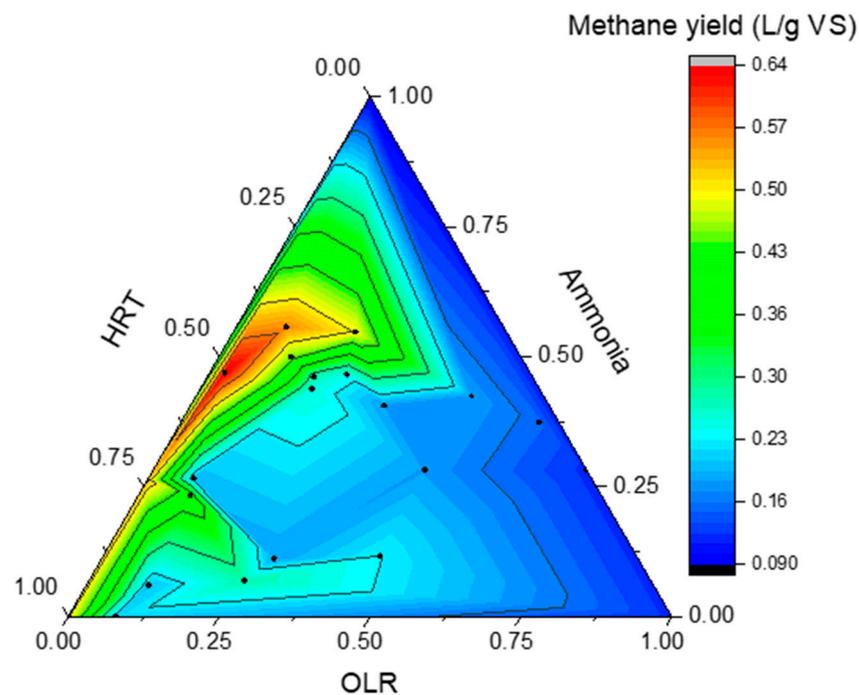


Figure 1. Representation of methane yield data reported in Table 1. Values in this graph were normalized to unity. Extremes were added to facilitate visualization, assuming the maximum yield values to be those reported in Table 1 when OLR is at the minimum or HRT at the maximum, and assuming a 50% inhibitory effect when considering the maximum ammonia content, based on the results reported by Poggi-Varaldo et al. [31]. Data points are represented as black spots in the diagram.

A similar hypothesis was previously tested by Yan et al. [33] when studying the high-solid digestion of rice straw, including temperature as an additional parameter. On the other hand, Molinuevo-Salces et al. [34] studied the co-digestion of swine manure and vegetable processing wastes. They reported significant improvements in volatile solid removal when the content of vegetable wastes in the mixture was increased and also when the feed's solid content was lowered. Similarly, Habagil et al. [35] reported variations in biogas yields based on the organic load applied to the reactor and the C/N ratio, which was changed by altering the mixture proportion of food wastes and primary sludge. However, trying to set a specific C/N ratio for industrial digesters in an attempt to extrapolate results is not always easy. Large-scale digestion plants have to deal with the resources available in their surroundings all year round [18]. Therefore, these findings help predict the effect on plant performance based on the carbon proportion of the feed, although industrial plants still have to operate under optimum conditions based on the resources available to them.

There is a close relationship between the degradation rate attained and the residence time inside the digester. Thus, higher conversion rates may be achieved if solids can be recycled back to the digestion process. The return of organics back to the reactor guarantees a separation between HRT and solid residence time (SRT), leading to improved conversion

because complex organics spend a longer time in the system, favoring degradation. However, this option implies additional technological complexities in addition to a decrease in the volume of incoming fresh material, which also affects OLR. HS-AD is a way of increasing organic loading without affecting SRT. However, intermediary compounds and ammonia levels may reach inhibitory conditions.

Pastor-Poquet et al. [36] studied this process using the organic fraction of municipal solid wastes (OFMSW) as a substrate, with a content of 15% total solids (TS). These authors reported a 40% decrease in the methane yield when the ammonia concentration in the reactor reached a value of 2.3 g N-NH₃/kg. However, when adding an inert material to increase the solid concentration without modifying the nutrient content, the process could operate at greater solid values. The risk of acidification was only exacerbated when TS increased over 20%. The effect of adding an inert material is thus similar to diluting the media, but the phenomenon of water activity, in this case, imposes limits on the presence of solids inside the reactor.

Takashima and Yaguchi [37] studied the digestion of sewage sludge under HS-AD conditions and a thermophilic regimen. To prevent ammonia inhibitory problems, the authors implemented an ammonia stripping stage, applied to the digested sludge. The process also included the return of the digestate back to the reactor to allow complex material to remain inside for a higher retention time, thus attaining its complete degradation. The stripping of ammonia kept the values of total ammonia nitrogen (TAN) at 1720 mg N/L inside the reactor (below the 2500 mg N/L threshold, a value reported as inhibitory in this study), making it possible for the reactor to operate at an influent concentration of 9–10% total solids.

3. Solid-State Anaerobic Digestion (SS-AD)

Solid-state fermentation is another way of operating digestion technology. TS values are higher than those used in HS-AD and seem more suitable for treating agricultural residues and food wastes, due to the lower water demand. Agricultural wastes have an intrinsic capacity to act as a structuring agent during fermentation because of their high content of lignocellulosic material. Many agricultural residues are untreated or underutilized, creating climate change problems associated with the emission of greenhouse gases (GHG) during the uncontrolled degradation of this type of waste [38]. Solid-state fermentation has been successfully studied for producing enzymes, biosurfactants, proteins, and biofuels [39,40] and a great variety of valuable products [41–44]. Anaerobic digestion has also been evaluated under this configuration and the co-digestion of different residues, such as manure, food wastes, and agricultural wastes, has also been studied under SS-AD conditions [45–47].

3.1. Operating Conditions and Leachate Bed Configuration

Operating conditions, such as nutrient levels, the feedstock-to-inoculum ratio, pH, temperature, and mixing, need to be carefully controlled to ensure success under SS-AD [48]. Solid-phase fermentation can be operated under batch conditions with low operating costs and low maintenance requirements [49], but other operating modes such as continuous and multiple stages have been implemented [48]. Under batch conditions, reactors are loaded with the substrate, and therefore inoculation is provided with each load. This type of operation creates an uneven evolution of biogas because the methane production rate is very high at the beginning of the process when the reactor has just been loaded, but as the digestion proceeds, the gas evolution slows down until the digester is again reloaded [50]. Operating in a staggered mode with biogas storage is a way to attenuate this non-uniform production of gas. However, scale factors and higher installation costs may limit the applicability of these measures.

The recirculation of leachate or a liquid phase, rich in anaerobic microflora, is a common practice to allow for the redistribution of soluble compounds and microorganisms. However, as degradation and solubilization of the organic material take place, compaction

of the organic bed may be experienced if not enough structuring agent is introduced when initially loading the reactor [51]. The purpose behind adding a structuring agent is to create a porous media to favor liquid circulation and prevent the formation of preferential pathways, which may lead to localized acidification in inner reactor zones. The structuring agent also helps in reducing localized organic loading in the reactor, acting as a dilution media, but it may also exhibit the undesirable consequence of causing a severe decrease in the reactor working volume and thus compromising its main advantage regarding volumetric methane productivity. Lignocellulosic biomass, such as straw and wood chips, are suitable structuring materials.

The scarcity of water available for attaining microbial conversion creates an environment where VFAs build up and high ammonia levels are easily reached. Co-digestion in the presence of a structuring agent alleviates the excessive increase and accumulation of inhibitory compounds in the liquid phase and avoids preferential pathways for fluid circulation in these units. The lack of adequate mixing creates difficulties, associated with mass transfer limitations. However, attempts to perform mixing in a digestion bed with high viscosity result in disadvantages, as high installation and operating costs are derived from the increased energy demand. On the contrary, maintaining low mixing levels then leads to a longer time needed for completing the total degradation of organics [52]. For this reason, leachate circulating reactors are more attractive due to their lower energy demands and technological complexity.

The recalcitrant nature of lignin in relation to anaerobic microflora aids in creating a porous structure and serves as a support to sustain biomass growth. Leachate recirculation and the frequency of this operation have a significant effect on the stability of solid-phase digestion. Qian et al. [53] evaluated this type of process and indicated that recirculation contributed to the enhancement of hydrolysis and acidogenesis, thanks to an inoculating effect, and favoring mass transfer. However, these authors pointed out that when recirculation was excessive, then a negative outcome was observed because it then caused microbial biomass washout and VFA accumulation. Xing et al. [54] also evaluated the recirculation of leachate, treating lignocellulosic biomass (*Pennisetum hybrid*) as a substrate. They found a similar detriment in reactor performance when the frequency of recirculation was increased. However, in a later experiment, Qian et al. [55] reported that adding a solid inoculum when loading the solid-phase reactor allowed them to increase the level of leachate spraying, favoring digestion and almost doubling the specific methane yield (from 0.107 L CH₄/g VS, reported for the liquid-inoculated system, to reach a value of 0.184 L CH₄/g VS when the solid inoculum was added to the reactor; these values were reported for the treatment of a mixture of OFMSW and corn stover).

The separation of the digestion system into two stages, the first dedicated to the hydrolysis and acidogenesis of the feeding material and the second to the conversion into methane of the acidified liquor, is a way to overcome acidification problems and buffering accidental overloading. When applied to solid-phase systems, the first phase acts as a leachate bed reactor and the second one as a traditional CSTR or as an up-flow anaerobic sludge bed (UASB) system [56–58]. Liu and Liao [59] studied a two-stage process, with the first stage operating as a leachate bed reactor (LBR). These authors attained the conversion of the substrate in less than 6 days, with a 70.9% removal of VS from the leachate reactor. However, if the total mass of substrate loaded (10 kg) and the volume of the reactors are considered (70 L for the LBR and 35 L for the second methanogenic phase), the OLR applied would be equivalent to 1.5 g VS/Lr d—expressed in terms of the volume of the reactor—when the loading estimation is performed for a continuous system. Biogas production in the LBR displayed an evolution characterized by a peaking behavior immediately after the addition of the methanogenic leachate and a rapid decrease due to the excessive accumulation of VFAs. These authors also reported compaction to be a problem and hydrogen gas evolution was described during the initial recirculation stages, which indicates process imbalances.

Thaemngoen et al. [9] also studied a two-phase configuration system to treat Napier grass (*Pennisetum purpureum*), but in this case, compared the process performance with conventional wet digestion. Continuous leachate spraying from the methanogenic reactor promoted hydrolysis and prevented inhibitory conditions, but the system attained a methane yield for this substrate of 0.069 L CH₄/g VS, against a value obtained from biochemical methane potential (BMP) tests of 0.227 L CH₄/g VS and 0.158 L CH₄/g VS from a wet digestion reactor at an OLR of 4 g VS/Lr d.

The use of adapted anaerobic microflora is essential for improving the performance of this type of configuration, along with a suitable strategy for reducing the toxic levels of intermediates as they are produced in the course of the fermentation. Mahato et al. [60] studied this kind of two-phase system for treating a mixture of dairy cow manure and chicken manure at a temperature of 20 °C. The addition of solid inoculation and the continuous circulation of leachate from one reactor to the other allowed the authors to achieve a yield of 0.350 ± 0.110 L CH₄/g VS and prevented any effects regarding the accumulation of toxic intermediaries.

Configurations of alternating solid-phase reactors have also been evaluated, operating in staggered mode and using the freshly loaded reactor as the acidification bed, receiving the leachate from the reactor, close to finalizing the digestion process. This strategy, represented in Figure 2, allows for the removal of the VFAs generated at a much higher rate at the beginning of digestion. The irrigation of this leachate over mature reactors increases the biogas production rate during their final stages [61]. This type of process was first described by Chynoweth et al. [62] and Chugh et al. [63] as a way of improving the degradation rate of leachate bed reactors by applying leachate recirculation strategies.

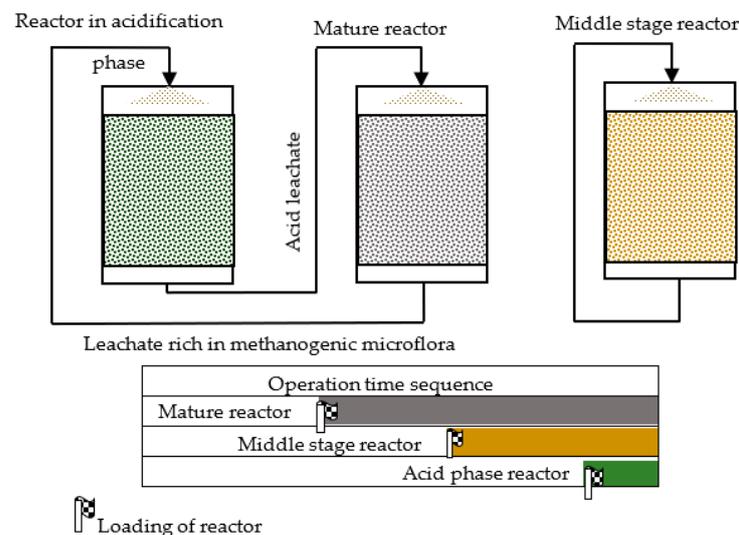


Figure 2. Schematic representation of a batch sequential solid-phase reactor with leachate irrigation as proposed by Chynoweth et al. [62] and Chugh et al. [63].

Other processes operating under similar configurations included mixed stabilization, where a first aerobic phase is introduced to increase hydrolysis performance [33]. This initial aeration phase aids in accelerating the hydrolysis stage of the process and reduces heating requirements, since the temperature is increased thanks to the short composting stage that takes place [64]. Gómez et al. [65] evaluated the stabilization attained under different solid-phase processes, some of which considered leachate bed configurations. In this study, the authors reported a rapid transition from the initial aerobic state to the anaerobic phase, attained through the spraying of anaerobic leachate. However, one of the major problems of these systems is the appearance of irregular zones where high VFA concentration may be found, preventing the further hydrolysis of the substrate, and pocket zones where methanogenic microflora may find protection [66]. This distribution may be

seen as a disadvantage since, in general terms, it will slow down the degradation of the substrate. Nevertheless, it can also be interpreted as a way of protecting the anaerobic biomass from a complete cessation of activity.

3.2. Commercial Technologies for SS-AD

Commercial technologies following the principle of the leachate bed configuration are currently available. The Bekon process, developed by BEKON GmbH, Unterföhring, Germany [67], is a single-step fermentation process using a garage-shaped fermenter. The inoculation of the system is carried out using previously digested material. A side-percolating fermenter contains leachate that is sprayed over the top of the fermenter. The Gicon process, developed by GICON Holding GmbH, Dresden, Germany [68], is a process using the leachate bed configuration but operating without an initial inoculation with a digested bed. Percolating reactors are, in this case, responsible for adding the amount of microorganisms needed to complete the process. The combi-buffer tank and the methanogenic reactor, containing a packed bed, offer unique characteristics for process stability, creating optimal hydrolysis and methanogenic conditions. A similar process is the BIOFerm™ dry fermentation technology (by BIOFerm Energy Systems, Inc., Madison, WI, USA), where material remains for 28 days in the solid reactor [69]. Its process is characterized by simplicity in operation, similarly to the previous batch technologies, but with the added advantage of being optimized in the use of heat for keeping the leachate bed reactor at the desired temperature. Figure 3 presents a graphical representation of the main features of these three commercial processes. Other commercially available processes have been reviewed by André et al. [20] and Fu et al. [49].

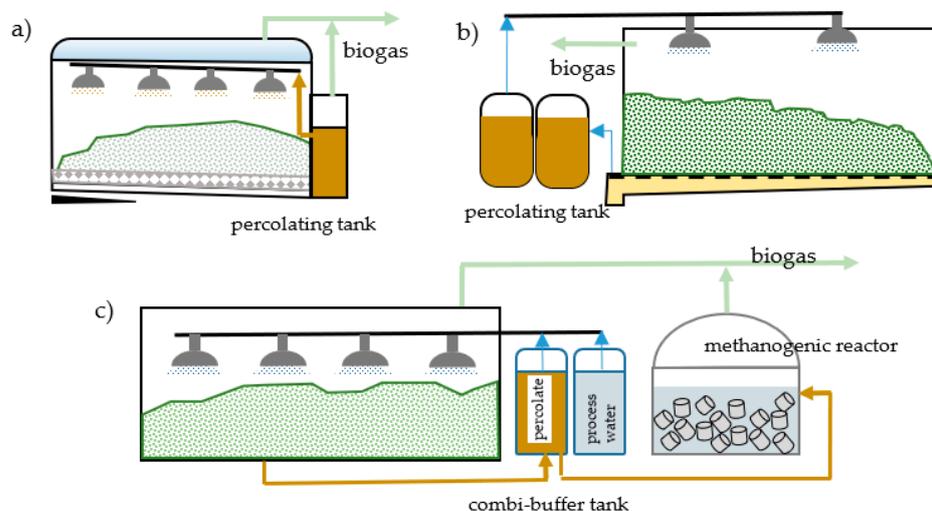


Figure 3. Schematic representation of leachate percolating processes at a large scale. (a) Bekon process, (b) BIOFerm™ system, (c) Gicon® process. Adapted from Fu et al., 2018 [49].

Two commercial solid-phase digestion processes with a large treatment capacity are Dranco (Organic Waste System (OWS), Gent, Belgium) [70] and Valorga® (VALORGA INTERNATIONAL, subsidiary of URBASER SA, Montpellier, France) [71], which are capable of dealing with an OLR greater than 10 g VS/Lr d under continuous operation and solid contents between 20–60% [72]. Kompogas® (Hitachi Zosen Inova, Zurich, Switzerland) [73] can also work under continuous operation, but lower loads are admitted. However, the thermophilic conditions set in this process allow digestion to be completed in around 14 days. In contrast with wet digestion, some of these dry systems lack internal mixing, and the incoming substrate and digestate are mixed prior to feeding the reactor [74].

The biogas yields obtained under solid-state conditions are lower than those from wet digestion systems. The increase in solid content causes a decrease in the biogas yield [75]. This fact was demonstrated by Li et al. [76] when evaluating the co-digestion of corn stover

and chicken manure under different configurations, that is, wet, high-solid, and solid-state digestion. These authors tested mixtures of substrates, but in general, the wet digestion system (at 5.1–5.6%TS) achieved higher methane yields than any of the other experimental set-ups working at higher solid contents. The methane yield was reported to be 0.219 L/g VS added for the wet system, whereas this value decreased to 0.208 L CH₄/g VS in the system with a high-solid content (10.1–11.2%TS) and further decreased to 0.148 L CH₄/g VS when evaluating solid-state digestion (20.1–22.4%TS). In addition, the optimum mixture composition for obtaining the highest methane yield was different for solid-state digestion, with a proportion of 1:1 (VS basis, corn stover/chicken manure), whereas for the other two digestion systems, this proportion was found to be 3:1.

A similar result was obtained by Ajayi-Banji et al. [45] when evaluating SS-AD using corn stover and dairy manure. These authors also reported the better performance of high-solid digestion systems when reducing the C/N ratio of the mixture, with these systems favoring the alkalinity and pH values of the reactor leachate. The effect of solid content should be considered when evaluating biochemical methane potential tests, since yields will be affected by the solid concentration used during the assay, in addition to the inoculum-to-substrate ratio (ISR) parameter. Holliger et al. [77] reported that biogas yields obtained from BMP tests compared well with those from large-scale digestion plants under wet and dry conditions (evaluating the Kompogas[®] process). However, the authors gave no indications regarding the solid content at which these tests were carried out. Studies performed by Wang et al. [78] and Molinuevo-Salces et al. [34] indicated the relevance of several parameters when evaluating biogas yields, reporting that C/N and substrate loading were also factors affecting the final cumulative production in addition to ISR and feed composition (co-digestion mixture percentage).

Kim et al. [79] studied the effect of moisture content in SS-AD using a bedding material composed of sawdust collected after 2–3 months of being used as cattle bedding. These authors evaluated this material as a substrate, which had a solid content between 17% and 30%. Although the values of methane yield reported were low for all cases tested, the system with a higher solid content presented a methane yield that was 29% lower than that at a TS content of 17%, thus corroborating the adverse effect associated with an extremely low water content. In solid-state fermentation, water activity (a_w) has a determinant influence on microbial activity, having a fundamental role in the mass transfer of water and solutes across microbial cells [80]. Therefore, there is clear evidence on the limits imposed regarding the levels of inhibitory components and the water content of the system, and their removal from the liquid phase is a necessity during SS-AD to avoid excessive toxic effects. The strategy proposed by Takashima and Yaguchi [37] of introducing an ammonia-stripping stage in HS-AD systems treating sewage sludge seems reasonable and leads to an expectation of success in digestion systems operating at even higher solid contents. Indeed, this is what Farrow et al. [81] intended when digesting poultry manure under a solid-phase configuration using struvite precipitation with pH controlled at around 7.0 during the ammonia removal stage to avoid adverse effects on the microbial biomass. This strategy allowed biogas to increase by about 30% under batch conditions and by nearly 235% when operating under semi-continuous conditions, reporting biogas yields of 0.420 ± 0.050 L/g VS added. However, the OLR was extremely low for an SS-AD system (OLR of 1.5 g VS/Lr d) and it should be added that they also experienced a decrease in the biogas yield with the increase in OLR.

The performance of high-solid and solid-state digestion systems needs to be increased by reducing the levels of the different digestion intermediaries and end-products that can exert toxicity over the microbial biomass. Nevertheless, given the low water level of this type of configuration, other options should also be considered, as these may improve the tolerance of anaerobic microflora or provide protective sites that may aid in temporarily removing inhibitory compounds. This may be attained by adding active compounds that help the microflora survive under these extreme conditions or provide alternative degradation routes.

4. The Effect of Adsorbents and Materials in Accelerating Anaerobic Degradation

Agricultural residues with a high lignocellulosic content and a low moisture content may represent an excellent potential energy resource [82] to produce biogas as a valuable fuel if the proper conversion can be attained at a reasonable cost under a solid-state configuration. In addition, co-digestion with manures may take advantage of the synergistic effects reported by several authors [83–86], particularly when the solid content of the system is increased. However, some difficulties still need to be solved, such as the higher degradation time needed, the high inoculation rate required to start up this process, and the low degradability of lignocellulosic biomass. The presence of lignocellulosic material in agricultural wastes serves as a structuring agent, avoiding compaction, but reduces the methane yield. However, any attempt to increase biodegradability will lead to mass transfer limitations and non-uniform liquid circulation through the bed.

The addition of adsorbents and carbon conductive materials to anaerobic reactors has been evaluated with success to decrease the impact of inhibitory compounds [87,88]. Adding this type of supplement to digestion allows for the enhancement of biogas productivity without greatly affecting the energy demands of the process [89]. The use of biochar derived from the thermal processing of lignocellulosic biomass in digestion systems has awakened interest among the scientific community, given its proven benefits regarding the mitigation of the negative effects of VFA and ammonia [90,91]. Other materials, such as zeolites, activated carbon and various adsorbents (kaolin, silica gel, polyvinyl alcohol, among others) have also provided benefits in biogas production [92–95] but the costs associated with these initiatives need to be carefully evaluated.

These different strategies may be useful in alleviating some of the difficulties found in solid-phase digestion and HS-AD. Petracchini et al. [96] studied HS-AD of food waste and cow manure using natural zeolite to prevent the effect of inhibitory compounds. These authors reported a biogas yield of 0.680–0.920 L/g VS. Calabrò et al. [97] evaluated the digestion of sewage sludge in the presence of high values of VFAs, analyzing the effect of different supplements, testing granular activated carbon (GAC), aluminum powder, granular iron, and steel scrap powder. Successful results were obtained when adding GAC and aluminum particles. Cuetos et al. [98] also demonstrated the benefits of using GAC when digesting blood obtained from poultry slaughterhouses, reporting that the digestion of this single substrate was not possible unless this material was added as a supplement. Recent research activities carried out by Dastyar et al. [99] evaluated a leachate bed recirculating reactor for solid-phase digestion with the addition of powdered activated carbon. However, the increase in the biomethane yield was just 17%, compared with the control system, which was also digesting the organic fraction of municipal solid wastes. Given the high price of activated carbon, low-cost adsorbents or strategies for increasing the benefits obtained should be considered to allow the industrial implementation of these solutions.

The mechanism of direct interspecies electron transfer (DIET) has been frequently proposed to explain the better performance of anaerobic digestion when carbon conductive materials are supplemented [100,101]. The enhancement is explained by the availability of a faster degradation route for the conversion of VFA [102–104], which is possible due to the prevalence of microbial species that become dominant due to the presence of materials that favor electron transport. Guo et al. [105] demonstrated the efficacy of adding GAC or magnetite on propionate degradation. These compounds favor the dominance of a syntrophic consortium by creating a DIET environment.

The addition of nanoparticles to digestion systems has recently demonstrated benefits in biogas production and the reduction of conversion times. The mechanism and effects of nanoparticles in anaerobic digestion have been reviewed by Abdelsalam et al. [106] and Faisal et al. [107]. Nanoparticles cause microbial activity stimulation based on the higher bio-availability of metal components essential for enzymatic reactions, thus enhancing cellular growth. Nanoparticles of iron oxide and zero-valent iron enhance interspecies hydrogen transfer and direct interspecies electron transfer, explaining the excellent results

obtained when they are supplemented into digestion systems [108]. Other metals (Cu, Co, Ag, Ni) and metal oxides have also been studied as supplements in anaerobic digestion in the form of nanoparticles [109–112]. Nanomaterials, in general, may become a useful ally in promoting substrate degradation due to their unique characteristics such as their high surface area, high reactivity, and specificity, and their increased number of active sites [113]. As observed in Table 2, there is a wide variety of reports available in the literature on the benefits associated with the addition of conductive materials and adsorbents.

Table 2. Results reported in the literature regarding methane enhancement when different types of supplements are added for the prevention of inhibitory conditions or the favoring of microbial performance.

Supplement	Substrate	Benefits	Biogas Yield Increase	Reference
Carbon conductive materials				
Biochar	Food wastes	Reduce digestion lag phase	33–275%	[114]
	Food waste components	Increase process's alkalinity, CO ₂ removal	77.5–98.1% (methane yield)	[115]
		Reduce digestion lag phase	4.74 times higher	[116]
	Citrus wastes	Reduce digestion lag phase, favored co-culture formation	56%	[117]
	Animal carcasses	Faster degradation of lipids and proteins	24%	[103]
	Brewer's spent grain	No enhancement clear	High variability in results	[118]
Hydrochar	Fruit wastes	Reduced VFA formation	13–27%	[119]
	Waste-activated sludge	Enhancement of acetoclastic pathway	46.9%	[120]
	glucose	Enhanced hydrogenotrophic methanogenesis	15–29%	[121]
	Graphite	Waste-activated sludge	Enhancement of acetoclastic pathway	38.3%
Adsorbents				
Biochar + zeolite	Cassava wastewater + livestock manure	Reduce digestion lag phase	No enhancement clear	[88]
Mg-zeolite, Co-zeolite, Ni-zeolite	Piggery waste	Increased biodegradability	8.5 times higher (Mg-zeolite), 4.4 (Co-zeolite), 2.8 (Ni-zeolite)	[122]
Zeolite	poultry slaughterhouse waste	Reduce ammonia concentration in digesters	15%	[95]
Bentonite	Waste activated sludge + kitchen waste	Reduce digestion lag phase	Two–threefold increase	[123]
Eggshell and lignite-modified zeolite (ELMZ)	Synthetic media evaluating high-ammonia conditions	Increase degradation rate	7-fold higher when compared with natural zeolite system	[124]
Granular activated carbon (GAC)	Orange peel wastes	Good process stability	65%	[125]
Sorghum-based activated carbon	Food waste + sewage sludge	Ammonia and TVFA concentrations were reduced	35%	[87]
Zero-valent iron (ZVI) + activated carbon	Waste-activated sludge	Increase in methane content, greater removal of organics	37.6%	[126]
Aluminum powder, pectin, gelatin, silica gel, bentonite, powdered activated charcoal	Cattle dung, poultry waste, cheese whey (2:1:3, w/w dry weight basis)	Adsorbents provide a site for anaerobic reaction to take place; 17% greater methane content	Twofold gas enhancement	[127]
Addition of nanoparticles				
Zero-valent iron (ZVI) Fe ₃ O ₄ nanoparticles	Food waste and waste activated sludge	Higher biodegradability	50% with Fe ₃ O ₄ No significant effect with ZVI	[128]
Co, Ni nanoparticles	Animal manure	Reduce lag phase and degradation time	1.64–1.74 times increase	[129]
Metal oxide nanoparticles (Fe ₃ O ₄ , MgO) and Ni, Co nanoparticles	Microalgal biomass	Increase in biogas production rate	8–28%	[109]
Fe ₃ O ₄ nanoparticle + microwave pretreatment	Green algae (<i>Enteromorpha</i>)	Increase in biogas production rate	54% ¹	[130]
Graphene oxide nanoparticles	Pre-treated slurry mixed with wheat straw	Increase in volumetric production at 40 days HRT	1.74–2.54 times increase	[131]

¹ Estimated from digitized graph reported in [130].

Casals et al. [132] reported a threefold increase in methane production when supplementing iron nanoparticles (NPs). Abdelwahab et al. [133] studied the digestion of cattle manure and obtained a biogas yield of 0.953 L/g VS when evaluating a concentration of 15 mg/L of (Fe) NPs against a value of 0.589 L/g VS obtained from the control experiments. Not only was the biogas yield enhanced, but the presence of these particles also favored a lower production of H₂S, which is of great relevance regarding subsequent biogas up-grading operations. Similarly, Farghali et al. [134] studied the addition of iron oxide (Fe₂O₃) and titanium dioxide (TiO₂) nanoparticles, reporting a twofold increase in biogas yields and a decrease in H₂S production. The addition of magnetite NPs was studied by Ali et al. [135] and Zhong et al. [136], with the latter indicating that the presence of these particles was probably responsible for accelerating the transfer of electrons from acid oxidizers to syntrophic methanogenesis, stimulating acid oxidizers to degrade acetate

into H_2/CO_2 , and finally to facilitate methane production. These reports open a new line of research completely disrupting the current efficiency of digestion plants, improving performance, and offering a completely radical change in the valorization of biogas. However, other factors—more than just economic criteria and bioenergy production—must also be evaluated when considering organic waste treatment. Sociocultural ideas, environmental impacts associated with this technology, and local knowledge may appear as important constraints [137], necessitating careful assessment to avoid causing a negative perception in local communities.

5. Temperature and Digestion Performance

Temperature is a crucial parameter for increasing the degradation rate. Psychrophilic conditions refer to systems working at temperatures lower than 20 °C, mesophilic conditions range between 20 °C and 45 °C, and thermophilic conditions have temperatures higher than 45 °C [138]. Any increase in temperature will translate into a greater biogas production rate, and therefore it is reasonable to assume that ideal operation should be based on optimum temperature conditions. However, this is not always possible since capital investment and operating costs are also parameters that greatly influence plant profitability. Thus, operation at low temperatures has been studied to determine the decrease produced in process performance and evaluate ranges of feasible operation [139]. The absence of a heating system to reduce operating costs also leads to variable performance due to daily temperature variations, which may cause process instabilities [140], and extremely low activities in the winter season.

SS-AD has been tested at temperatures below 34 °C. Since the main advantage of this technology is its simplicity, the installation of a heating system would add unnecessary operating costs. Avoiding these additional costs is vital if this technology is extensively applied in developing countries and/or tropical countries where excessive low ambient temperatures are not experienced. Ghosh [141] evaluated the fermentation of solid wastes around 25 °C, obtaining a yield of 0.26 L CH_4 /g VS added, thus proving the suitability of this process even at this temperature. Operating at lower temperatures to establish optimum conditions for low-cost digestion systems is needed.

Psychrophilic digestion has been studied by different authors, reporting lower biogas yields [142,143] and solid accumulation [144], but successful experiences have also been described, with gas yields similar to those obtained at higher temperatures, indicating that the process was not significantly affected by the increase in the OLR, as would be expected [145,146]. These reports are important as many small-scale digesters operate under this regimen. When the performance of these systems is analyzed, better yields are obtained than those expected from control laboratory conditions. This is probably explained by the well-established consortium attained after an extended operation time in industrial operating reactors [147]. Zhao et al. [148] studied digestion performance at 4 °C, indicating that the maximum treatment capacity was set at 4.33 g VS/Lr d of OLR. Therefore, low-temperature operating digestion systems may become a low-cost solution for the operation of decentralized reactors with a treatment capacity equivalent to that of more complex mesophilic and thermophilic reactors.

However, it is undeniable that increasing the temperature of the process affects reaction rates; therefore, to speed up biological degradation, the temperature should be increased. Moving from a mesophilic to a thermophilic regimen has been implemented to improve the treatment capacity of the reactor and thus productivity. Thermophilic conditions allow higher degradation rates, thus achieving a greater capacity for treating organics and attaining higher pathogen destruction [14]. A temperature rise from mesophilic to thermophilic conditions reduces the required volume of the digester and significantly decreases capital investment costs [149]. This feature translates into a significant increase in the treatment capacity of the plant for reactors that are already operating at lower temperatures but also result in a higher energy demand. The feed needs to be heated up to the desired thermophilic conditions, requiring a greater amount of energy, and this

demand is accentuated in the winter season. Thermal losses are also higher due to the greater temperature gradient associated with the process and the ambient temperature, making insulation crucial to avoid excessive energy losses.

The biogas yields for mesophilic and thermophilic systems have been reported to be similar, but some other authors have found greater yields when working at higher temperatures. Table 3 lists different biogas yields obtained under mesophilic and thermophilic conditions using BMP tests. To avoid the effect of inoculation and the characteristics of substrates, the studies listed in this table were those evaluating both conditions. There is great diversity in the results, but in general terms, the increase in temperature improves the degradation rate and requires less time to complete full substrate conversion. Thus, Kafle et al. [150] reported a value of k (first-order kinetic constant) of 0.033 1/d when evaluating the mesophilic digestion of food wastes. This value was increased to 0.075 1/d with the temperature rise to a thermophilic regimen (data obtained from BMP at a feed to microorganisms (F/M) ratio of 1, value expressed in terms of VS), which is interpreted as a higher hydrolysis rate, leading to a lower digestion time needed to complete the process. Ge et al. [151] evaluated the effect of temperature on the digestion of cellulose and reported an increase of 1.5 times the hydrolysis coefficient per each temperature increase of 10 °C.

Table 3. Biogas yields are reported in the literature. Data were obtained from different authors under mesophilic and thermophilic conditions, using biomethane potential (BMP) tests.

Substrate	Methane Yield (L CH ₄ /g VS)		Reference
	Mesophilic	Thermophilic	
Cow manure	0.120	0.120	[152]
Maize silage	0.400	0.550	[152]
Newspaper	0.046–0.061	0.077	[153]
Food wastes (F/M = 3) ¹	0.114 ²	0.700	[154]
Food wastes (F/M = 0.25–1) ¹	0.480–0.530	0.650–0.740	[155]
Chinese cabbage waste (F/M = 0.5–2.0) ²	0.591–0.677	0.434–0.639	[150]
Poultry slaughterhouse waste (intestine content)	0.610	0.675	[94]
Poultry feathers ²	0.200	0.276	[156]
Sewage sludge + fat ²	0.680	0.490	[156]
Cheese whey	0.304	0.160	[157]
Cattle manure	0.234	0.159	[158]
Maize straw silage ²	0.105	0.114	[159]

¹ F/M: food-to-microorganism ratio. ² Data digitized from graph reported in reference.

In some cases, a greater biogas yield may be expected when changing from mesophilic to thermophilic conditions [160,161], but even with similar yields, benefits are still gained based on the lower degradation time. However, stability issues are of concern. Labatut et al. [162] experimentally evaluated cow manure and simulated food wastes, indicating greater robustness for the mesophilic system, whereas the thermophilic one marginally outperformed the lower temperature reactor. Gebreeyessus and Jenicek [163] reviewed the performance of different mesophilic and thermophilic reactors and concluded that even though it is difficult to make exact comparisons when studies from different sources are evaluated, mesophilic systems seemed to be preferable because there are fewer stability issues associated with this technology in regard to high levels of free ammonia and VFA. Additionally, concerns may also be raised about the quality of the digestate (higher VFA and ammonia content under thermophilic conditions) and operational issues regarding sludge odor and dewaterability [164].

Nielsen and Petersen [165] reported on experiments with large-scale thermophilic digesters (50–55 °C), indicating a higher demand for polymer in sludge dewatering operations. De Vrieze et al. [166] also evaluated large-scale thermophilic performance in WWTPs located in the Netherlands. These authors indicated variations in digestate quality based

on an increase in the nutrient content (nitrogen and phosphorus) of the digestate. Working under thermophilic conditions has led to higher VFA and ammonia levels in the reactor liquor [167–169], which negatively affected digestate quality. Therefore, a post-digestion stage at lower temperatures may seem adequate if the land application is the final disposal option of the digested material.

Solid-state fermentation finds a niche application in treating farm livestock wastes and agricultural wastes. Manures are characterized by a high content of nitrogen, leading to a higher release of ammonia nitrogen. If thermophilic conditions are used to increase the release of this compound, then an inhibitory environment is easily generated, leading to a lower level of degradation and a lower quality of the digestate. Yenigün and Demirel [170] found discrepancies in mesophilic and thermophilic digestion results when reviewing different scientific reports available in the literature. These authors indicated that free ammonia values might be behind the differences in performance reported by several authors. Thus, higher free ammonia values obtained under thermophilic conditions negatively affect process stability, leading to the wrong conclusion that higher temperatures create greater susceptibility to the anaerobic microflora.

The increase in temperature also leads to better process performance when an adequate adaptation of anaerobic microflora is provided. Given the higher risk of solid-phase digestion in accumulating inhibitors, the addition of adsorbents and compounds capable of promoting the fastest degradation routes, such as carbon conductive materials, nanoparticles, or the introduction of bio-electrodes into digestion systems, may seem suitable and adequate for the operation of high-solid-content reactors or solid-phase digestion systems under thermophilic regimens. These strategies may increase productivity and reduce reactor size without causing significant detriments in biogas yields, favorably affecting capital investment costs and plant economic feasibility. In addition, increasing the temperature favors the degradation of highly lignocellulosic materials such as grasses [171] and other agricultural wastes, in which anaerobic digestion finds wide applicability, but on the other hand, may increase the risk of compaction and uneven degradation. Wang et al. [90] demonstrated the greater capacity of thermophilic systems to operate under higher organic loading rates when biochar was added to the reactor, due to the improvement in VFA degradation. Other techniques, such as micro-aeration, wherein small amounts of air are introduced into an anaerobic digester, have been shown to enhance biogas production. This occurs by fostering the growth of facultative aerobic bacteria and enhancing the production of enzymes that participate in the degradation of complex polymers such as cellulose [172,173]. Therefore, combining these different operating methodologies may provide a suitable means of reducing hydrolysis limitations and the accumulation of toxic intermediaries.

A two-phase digestion system for the treatment of municipal solid waste, involving micro-aeration and GAC added as a supplement, was evaluated by Canul Bacab et al. [174], demonstrating the feasibility of this approach for attaining fast hydrolysis, reducing digestion time, and enhancing methane production. An initial aerobic phase, prior to digestion, was proposed as a pre-treatment to increase the hydrolysis of lignocellulosic material [175]. There is no need for this pre-treatment phase to last for several days, with the authors reporting that 12 h of micro-aeration seems to be enough to observe a digestion enhancement [176]. In this method, a low energy demand pre-treatment stage is introduced into the conventional digestion process, leading to lower digestion volumes and lower initial capital investment. Micro-aeration not only favors the degradation rate of complex particulates but also presents additional benefits linked to the removal of hydrogen sulfide [177].

Based on the difficulties associated with high-solid and solid-phase digestion, novel reactor configurations, capable of achieving high hydrolysis rates of complex materials and lignocellulosic biomass, are needed. Attempts are currently underway to reduce compaction problems, to guarantee homogenization, and to favor ammonia removal from the system, such as the cartridge operating reactors proposed by Yang et al. [178], thus avoiding biomass floating problems and discharging issues [179]. New configurations

should consider digestion enhancement by supplementing low-cost materials capable of increasing biogas yields and attaining high levels of removal of volatile solids.

6. Conclusions

Anaerobic digestion is a suitable technology for the treatment of organics. There is still a wide range of methods for optimizing the operating conditions and reactor configurations and thus increasing treatment capacity and biogas yield. High-hydrolysis-rate reactors operating under solid phase configurations and/or high-solid digestion systems need to be developed. These reactors should maintain high biogas production rates and avoid inhibitory problems associated with the accumulation of intermediaries.

Novel configurations working under thermophilic conditions, without suffering from the problem of ammonia or VFA build-up, should be developed for anaerobic digestion to be considered a relevant technology for bioenergy production. The valorization of wastes through the various applications of digestates may ensure that anaerobic digestion becomes an environmentally friendly alternative that is capable of increasing the circularity of different production cycles. However, it is also true that simplified reactors and lower investment costs are necessary. Otherwise, digestion may not be able to become a key player in the new circular economy model.

The addition of supplements such as adsorbents, carbon conductive materials, and nanoparticles to anaerobic digestion may enhance reactor performance. However, other effects related to the presence of these components when the digestate is used in land applications should also be evaluated.

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