

Review

Data Driven Modelling and Control Strategies to Improve Biogas Quality and Production from High Solids Anaerobic Digestion: A Mini Review

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Abstract: Anaerobic Digestion (AD) is one of the oldest processes for producing biofuels from organic waste. Approximately 180 years have passed since the construction of the first modern plant, however, large prospects for improvement are still feasible, especially in regards to the quality and uniformity of the biogas produced. This work focalizes on the main quality issues and the available post-production treatment processes for biogas; subsequently, a mini-review on data-driven models and control strategies for biogas and bio-methane production plants is presented. Attention is focused on High Solids Anaerobic Digesters (HSADs), since these reactors present many interesting advantages, including a high number of operating variables which enable process optimization, high methane concentration in exit, reduced reactor volume and low water requirements. HSADs are the reactors with which Europe is aiming to rapidly increase the production of biogas and bio-methane, in order to carry out de-carbonization and reduce dependence on external methane imports. Crucial points for achieving these objectives include qualitative leaps in process operation and management, which, contrary to current practice in existing plants, require a significant increase in process automation, with control of product quality and reduction of stops due to death of bacteria at changing process parameters (such as temperature and pH). The most significant papers related to biogas quality, data-driven models and control strategies are briefly analyzed.

Keywords: High Solids Anaerobic Digesters; biogas quality; operating modes; data-driven models; control strategies



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1. Introduction

Biogas and bio-methane are accessible sources of renewable energy and can greatly contribute to de-carbonization of the gas sector, making renewable gases available for heating, transport and industry. Every year, the European Biogas Association (EBA, www.europeanbiogas.eu, (accessed on 30 October 2022)) publishes its enhanced edition of the EBA Statistical Report about developments of biogas and bio-methane production and market in Europe, as well as potential growth forecasts for the coming years. It also investigates the state of play of 19 national markets in Europe. EBA reports that today in Europe, there are about 20 thousand plants, including biogas, mixtures of biogas including methane and bio-methane. According to EBA forecasts, bio-methane could cover up to 40% of the total European gas consumption by 2050, with a production of one thousand TWh. Moreover, the actual situation in Europe, due to the recent Russia-Ukraine war, will push this growth even further in 2022 and 2023 in order to cover at least 30% of European gas consumption in 2030.

In 2019, the sector produced 167 TWh (15.8 billion cubic meters) of biogas and 26 TWh (2.43 billion cubic meters) of bio-methane. At the end of 2019, a total of 18,943 biogas plants and 725 bio-methane plants were operating across Europe. In 2018, there were 483 European bio-methane plants; this increased to 729 in 2020, with total production

reaching 32 TWh. In October 2021, Europe had 1023 bio-methane production plants and production increased 40% from 2020.

Among the European countries, France, Italy and Denmark have the highest growth rate of new plants. In 2020, 11 new plants came into operation in Italy and in 2021, there were about 20,000 production plants between biogas and bio-methane. The overall production capacity of bio-methane in Italy is 25,445 cubic meters per hour, equal to approximately 220 million cubic meters per year.

The production of biogas and bio-methane in Europe mainly comes from Anaerobic Digestion (AD) of animal waste and manure and organic and green urban solid waste [1]. A great potential for expansion is the anaerobic digestion of organic sludge from wastewater treatment plants [2,3]. Conversely, agriculture is the main type of feedstock used in other countries, including China [4]. An interesting comparison between biogas produced in Europe and China is reported in [5].

The potential expansion of biogas and bio-methane is significant, as the technology is easily available and widely mature; as far as Italy is concerned, bio-methane generated by the treatment of the organic waste fraction obtained from agricultural and animal waste could be tripled in a short time. In a few years, between 8 and 10 billion cubic meters of bio-methane and biogas could be produced in Italy, replacing a substantial percentage of methane imports.

If this expansion potential is exploited to the fullest, investment in new plants will increase considerably in Italy; however, from this arises the need to produce high quality biogas and bio-methane, so that it can be used immediately for production of heat and for the needs of the industrial sector. There are many types of AD processes available today, and they are optimized according to the type of feed (agricultural, livestock sewage, urban organic waste from separate municipal solid waste (MSW) collection, sludge from urban wastewater treatment plants). The choice between the different processes also depends on other factors, such as the availability of space and water resources. In recent years, operating with a high content of suspended solids and dry type AD (total solids (TS) content greater than 20%) is preferred to wet AD [6,7]. Finally, although the anaerobic digestion technology dates back to the mid-19th century, the production plants are not highly automated. The reasons are multifactorial: on the one hand, it is difficult to develop robust control systems due to the high uncertainty of feeding conditions (typically the concentrations of incoming organic substances vary greatly) and the lack of fast reliable predictive mathematical models; on the other hand, to improve the quality of the biogas produced, separate process units downstream of the production plant are preferred (by separation, treatment and purification of the biogas). High Solids Anaerobic Digesters (HSADs) allow the operation of feeds with high concentrations of suspended solids; moreover, it is possible to control different outputs due to a high number of operating variables involved and a more rapid reactor response to input variations, compared to that of Low Solids Anaerobic Digesters (LSAD), as discussed by Fagbohunge et al. [8] and Di Capua et al. [9].

Many review papers have been published on AD. Almost all of them focus on the type of feedstock utilized. Refs [10–13] discuss dry AD for organic waste treatment; Refs [14–16] present reviews on food waste anaerobic digestion; Refs [17,18] analyze the solid-state anaerobic digestion of lignocellulosic biomass. Moreover, Yao et al. [19] review the technological advances of AD to treat livestock manure; Momavez et al. [20] discuss dry AD fed with industrial crops waste; Refs [21,22] focalize on AD for treating chicken manure and straw /corn stover, respectively.

Some review papers can be classified in terms of biogas productivity, including work by [11,23–25], or in terms of AD potential for energy recover, such as papers by [26] about dry AD and work by [27] on high-rate AD.

Furthermore, a few authors focalize on general technological aspects to improve production [28], or on particular technologies such as biochar-mediated AD discussed in [29], micro-aerated AD analyzed in [30] and digestate management for HSADs reviewed in [31].

Only a very few review papers are dedicated to sensors [32], monitoring [33] and instrumentation and control [34] of AD processes. A recent review on the use of machine learning for AD can be found in [35].

This mini-review focuses on two main aspects of biogas and bio-methane production by AD: the quality of the product and the methodologies for process control based on data-driven models. The choice of data-driven models is preferred since they can be implemented more simply than physically based models, and they are tailored for control purposes. Moreover, they usually require a reduced number of measured variables for their identification, which is an advantage if models are proposed for improving both production and quality in existing plants. Conversely, models based on first principles require knowledge of a lot of chemical-physical parameters, whose estimation with low uncertainty is practically impossible in short times, due to the many phenomena involved (chemical reactions, biological reactions, mass and energy transport in multiphase systems). Moreover, if dynamic models are required for predictive control, the robustness of data-driven dynamic models is often better than that obtained with physically-based dynamic models, even if written in terms of state-space equations [36]. Finally, data-driven models can be constructed using data acquired during operation of existing plants, without specific knowledge about the chemistry and physics of the process, and using the existing sensors on the plant, or at least, by installing a few new ones.

This work analyzes the main possibilities to improve existing HSAD plants and primarily focuses on:

- the biogas quality requirements, and main treatment processes to obtain high-quality biogas;
- the data-driven mathematical models tailored for process optimization;
- the control strategies.

We used a data-prospection methodology, finding 250 papers using combination of ten specific keywords. After screening procedures, 148 papers were selected and organized into three main categories: biogas quality and treatment, data-driven models, control strategies. From these papers a total of 95 papers are evaluated and discussed in this mini-review.

2. Biogas Quality

2.1. Biogas Composition

Biogas is the useful product from anaerobic digestion; therefore, the whole process must be conducted in such a way as to maximize both the conversion (degradation) of the biomass and the methane yield, compatibly with the type of biomass used. The optimization of the conversion should be the primary objective in case the biomass is constituted by organic waste (MSW, manure). Even in the case of continuous or semi-continuous processes, both the flowrate and the composition of the biogas in exit from HSADs can present high oscillations. Flowrate variations can be around $\pm 40\%$, while concentration of methane in the biogas is usually between 50% and 65%. The variations in output of biogas flowrate and CH_4 concentration are inversely proportional. During the loading phase in semi-continuous HSADs, there is a large biogas production with low methane content, while in the aftermath of the loading phase, output of biogas decreases and the concentration of methane increases.

These variations are due to the different degradation kinetics of the different components inside the organic matter, and in single-stage processes, they are accentuated by the fact that the four main phases (hydrolysis, acidogenesis, acetogenesis, methanogenesis) of anaerobic digestion all take place within the same reactor. Single-stage HSADs are usually optimized, in terms of variables and process parameters, under intermediate conditions amongst the different phases, or are set to the best conditions for microorganisms responsible for the first three phases. From the analysis of both scientific bibliography and specific documentation of the companies that propose processes for the production of biogas from HSADs at industrial scale, the composition of a typical biogas is reported in Table 1 (in agreement to that reported in [37,38]).

Table 1. Typical composition of biogas from HSADs.

Typical Composition of Biogas [%]	
CH ₄	50–65%
CO ₂	35–45%
H ₂ O	0–5%
H ₂ S	0.02–0.2%
Siloxanes	Traces
N ₂	Traces
H ₂	Traces
O ₂	Traces

The quality of the biogas produced at industrial scale in plants that use MSW is quite similar to that of a landfill gas; therefore, a purification is always necessary before sending biogas for energy recovery. By using selected biomass the quality of biogas can increase, but purification is necessary both for environmental impact reasons and to avoid damaging the systems used for energy production. The purification processes must be more or less driven according to the final use of biogas (internal combustion engines, gas turbines, fuel cells, etc.). For example, purification is less stringent for biogas use in internal combustion engines than its use in gas turbines, or even in Polymer Electrolyte Membrane (PEMFC) or Molten Carbonate Fuel Cells (MCFCs) with integrated Methane Steam Reformers (MSRs).

The feasibility and methods adopted for energy production through the use of biogas from HSADs are strongly influenced by the concentration of methane in the mixture, i.e., the recovery process is economically advantageous only if methane volumetric concentration exceeds 50–55%, and by costs of biogas purification. In addition to the main components, biogas can contain more than 140 additional trace chemicals that reach a total concentration of about 2000 mg/m³, i.e., about 0.15% volume.

In general, the presence of carbon dioxide and water causes the decrease of the LHV of the mixture, while substances such as hydrogen sulphide, oxygen, siloxanes, halogenated organic compounds and acids that may be present in traces act as corrosive agents, causing significant damage to the plant.

The main processes required to remove main pollutants are filtration, dehumidification, desulphurization and carbon dioxide removal. These processes can be carried out primarily in the following ways [39]:

(a) Filtration

At the exit of the digester, filtration (<10 mm) must be provided to eliminate liquid or solid particles which could be entrained by biogas. This simple system protects blowers or compressors that will be used to supply gas to subsequent units.

(b) Dehumidification

The dehumidification treatment is necessary because humidity, in which biogas is saturated, can condense inside the pipes, which changes temperature and/or pressure and causes malfunctions. The condensed humidity contains precipitates of harmful and corrosive substances. Dehumidification can be achieved by cooling the biogas.

(c) Desulphurization

The desulphurization of biogas containing sulfur compounds [40] at low concentrations can be carried out with different processes [41,42].

Absorption (scrubbing). This process is usually adopted for gases containing H₂S at medium concentrations and if the recovery of H₂S is advantageous. Water is used and CO₂ is also absorbed. Desulphurisation must take place before dehumidification. High volumes of water are required and a distillation process (reboiling) is adopted for H₂S recovery. Other solvents that allow physical absorption (methanol, ethers, glycol-polyethylene, propylene carbonate) can also be used and are also recoverable through thermal processes.

Solvents within which chemical absorption occurs can also be used, generally under conditions of pressure and ambient temperature. In such cases, any regeneration of the solution takes place by reoxidation. A very simple type of treatment consists of washing with a basic solution, which neutralizes H_2SO_4 (Dow Chemical Co., Midland, MI, USA, GTP-Merichem, Seaboard process). A subsequent acid washing phase allows the neutralization of the basic excess before the solution is discharged. The cost of reagents and wastewater treatment is usually relatively high if compared to the economic value of the purified product. Alternatively, oxidant compounds are added continuously to oxidize H_2S . Oxidants recommended include chlorine, sodium hypochlorite, calcium hypochlorite, hydrogen peroxide, ozone, sodium nitrite and potassium permanganate [43].

Treatment with solvents like monoethanolamine (MEA), diethanolamine (DEA) methyldietanolamine, (MDEA) and di-isopropanolamine (DIPA) in water solution leave a concentration of residual H_2S in the exiting biogas of the order of few ppmv. Finally, other liquid-based processes are Chemsweet[®] (Natco, Inc., Dehradun, India) using a zinc-oxide, or the absorption by tetra-n-butyl ammonium bromide (TBAB) semi-clathrate hydrate [44].

Physical adsorption at low temperature. Gas-solid adsorption is the most common process for the removal of H_2S and volatile compounds from biogas. The adsorbent can be regenerated by desorption at high temperature (TSA), low pressure (PSA), under vacuum (VPSA) or by stripping with inert gas. Molecular sieves such as zeolites, silicas or zinc oxide [45] with different pore sizes can be used; they also allow the adsorption of mercaptans (R-S-H) and CO_2 . The use of natural zeolites (clinoptilolite) in industrial processes has assumed considerable importance given the current availability at competitive prices compared to those of activated carbon. The use of zeolites supported with solvent films, e.g., triethanolamine [46] or N-methylpyrrolidone seems to be a promising process due to good regeneration.

However, the most widely used adsorbent still remains activated carbon, both in the form of very high porosity coal and in the form of molecular sieves. Activated carbon can be regenerated by heating with inert gas (steam or nitrogen). In industrial plants, activated carbon is not often regenerated when adopted for the adsorption of H_2S .

The choice between un-impregnated or impregnated activated carbon [47] depends on the quantities of H_2S to be removed, the acidity, the surface pH and the pressure drops [48]. Activated carbons impregnated with basic solutions (typically KOH and NaOH) are most commonly used for the purification of landfill biogas from H_2S . In the presence of alkaline compounds, the oxidation of H_2S involves the deposition of elemental sulfur on coal; this process is also facilitated by the water film present on the surface of the coal [49]. Impregnated coals are not usually regenerated [50]. Although these solutions involve a high removal efficiency, the use of alkaline solutions has some disadvantages, including: risk of autoignition due to an exothermic reaction between basic compounds and the CO_2 present in the biogas to be purified; the decrease in the yield in terms of active surface on the volume of adsorbent (due to occupation of the pores by the impregnating agent); the costs of the impregnating agent and its corrosive action. To increase efficiency, solutions have been proposed that involve the use of activated carbon impregnated with caustic and supported with metal oxides (zinc, copper, iron).

An important parameter to evaluate when choosing the adsorbent is the amount of water present in the biogas. Typically zeolites work better (due to H_2S removal efficiency) in dry biogas conditions, while activated carbon achieves higher yields in the presence of humid currents. The last variable to be taken into consideration in the choice of adsorbent concerns is in the compromise between regeneration and disposal, which can lead to landfilling or destruction. A possible advantage in the use of activated carbon is the possibility of disposal directly by thermal disruption.

Chemical adsorption at low temperature. It involves the use of solids, typically iron oxides and hydroxides, which are not typically regenerated. Some commercial solids include Sulfatreat 410-HP[®] [51], a non-toxic granular material containing Fe_2O_3 , Fe_3O_4 and an activator developed for the treatment of biogas with dimensional properties that guarantee

high external and internal diffusion and reaction kinetics [52]; IRON SPONGE [53], which is made from wood chips and supported by hydrated iron oxide, is proposed for the purification of landfill biogas and biogas from HSADs. IRON SPONGE requires a high degree of humidity in the biogas, which can be obtained by spraying water or with a stream of steam. The H₂S adsorption reaction also produces H₂O, which already contributes to keeping the filled bed moist. The reaction can take place in temperature ranging from 10 to 50 °C and is not influenced by pressure. Another solid material based on iron oxides is Sulfur-Rite (GTP-Merichem), whose removal yield seems to be higher than that of iron sponge systems. This adsorbent is used to remove H₂S at low concentrations since its cost is high. Adsorption at medium-high temperatures with zinc-oxide based adsorbent materials is processed at temperatures around 200–400 °C (adsorbents from the PURASPEC family) [54]. Mercury and chlorine-based compounds can also be removed with these materials [55].

Biological removal. Biofiltration consists of wet beds in which many microorganism species are capable of degrading sulfur compounds [56]. The final concentration of H₂S mainly depends on porosity, temperature, pH, humidity and H₂S initial concentration in the gas phase. Some of these processes are BIODESULF™ by ARCTECH (mixed microbial culture) BIO-SR by Dowa Mining Co, Vancouver, BC, Canada, (*Thiobacillus ferrooxidans* bacteria), THIOPAQ™ by Paques Biosystems, Montreal, QC, USA, (*Chlorobiaceae* and *Chromatiaceae*), UOP by Honeywell, Charlotte, NC, USA, [57]. UGN-BEKOM (H) system [58] is an interesting process with a combination of chemical and biological removal of hydrogen sulfide.

Carbon dioxide removal. In some cases it may be useful to carry out treatments to remove or reduce the CO₂ content, if aiming at increasing the concentration of methane in the biogas. The removal of CO₂ takes place either by absorption/stripping or with the use of semipermeable membranes. The process follows that of removing the H₂S.

2.2. Biogas Impurities

The problem that can arise from the use of biogas as a fuel for the production of electricity is that different components, even when present in traces, can damage the gas engines in which the fuel is used, as well as the heat exchangers and catalytic treatment systems of the exhausted gases located downstream. Among all the components, the most harmful are H₂S and the components that contain silicon, i.e., those belonging to the siloxane family. If any section of the plant in which the biogas flows is subjected to high temperature, and in the absence of a prior separation of the siloxanes and H₂S, a series of corrosion problems can severely limit the benefits derived from the use of biogas as an energy source [59]. During the combustion process, the H₂S reacts with halogenated components and oxygen also present in the biogas, forming corrosive acids such as H₂SO₄, HCl and HF.

The siloxanes, on the other hand, are converted into microcrystalline silica (SiO₂) with physical and chemical properties very similar to those of quartz, which stratifies uniformly and creates an abrasive layer on the entire gas-solid contact surface between the biogas and the internal walls of the system, assuming a thickness or a growing function of the time of use and of the flow rate of the evolving gas.

While the problems deriving from the development of corrosive gases can be solved with the adoption of highly resistant construction materials, those deriving from covering internal walls with siloxane layers can lead to corrosion of the surfaces with which they come into contact, causing overheating of the covered areas acting as thermal insulators, alterations in the instruments for measuring the parameters in the combustion chambers of gas engines and reduction in the potential for controlled ignition device, functioning like electrical insulators. They also damage all sorts of combustors, exchangers and internal combustion engines in each of their parts, including: tuber bundles, ignition devices, internal walls of the cylinder, pistons and intake and exhaust valves.

Removal of Siloxanes

The term “siloxanes” refers to a subset of silicones with linear or cyclic structure containing Si-O bonds with radicals surrounding the silicon atom, such as the methyl group, ethyl group and other organic functional groups. Siloxanes are not decomposed in processes that use activated sludge, but are preferably adsorbed on the EPS (Extracellular Polymeric Substances).

Different methods based on the separation by liquefaction of biogas are technologically feasible but excessively expensive from energetic and, therefore, economic point of view.

Since biogas contains a wide range of components (H_2S , siloxanes and organic) with concentrations that span several orders of magnitude, a rather high adsorption of both siloxanes and all other trace components must be achieved. Thus, one of the fundamental requirements that the adsorbent material must possess is a high adsorption capacity, so as to retain components present even in very low concentrations. However, given their strong reactivity as a consequence of a high surface energy, the active sites of the adsorbent material also adsorb water vapor and other polluting agents. The presence of other pollutants leads to a fast decrease in the useful life of the adsorbent, which is time needed to saturate all the micropores with the adsorbed substances. All the abatement processes currently adopted are effective in removing siloxanes from biogas; the most frequently used are those that provide the treatment of biogas with activated carbon, which obtains a physical adsorption of the siloxanes inside the micropores of these materials. Other potential adsorbents are zeolites, polymeric beads and silica gel [60].

In the most common adsorption units, activated carbon is used to reduce the siloxane content, but since these pollutants have a rather limited tendency to desorption, regeneration of the adsorbent bed cannot be achieved and these beds must be replaced regularly. Other adsorbent materials used for the removal of siloxanes from gas phases include molecular sieves, such as zeolites and polymeric beds.

Experimental studies have been conducted on the adsorption capacity of materials including polymeric extrusions and silica gel. Both show marked adsorption capacity, especially with respect to siloxane D5 (decamethylcyclopentasiloxane). Silica gel seems to be the material par excellence for biogas treatment as it allows a simultaneous separation of water (gas drying), in addition to the separation of polluting compounds. The regeneration efficiency for silica gel can be higher than 95% if brought to a temperature of 250 °C for twenty minutes. In similar studies on activated carbon, thermal regeneration was less efficient, even though it exhibits a siloxane adsorption efficiency of approximately 99.1%. The problem, therefore, still remains the cost.

Polymethyl siloxanes may be decomposed by producing methane and silica-like or silicates materials over metal oxides at high temperature [61].

Siloxanes can be also removed by absorption in liquid phase. Huppmann et al. [62] used tetradecane as a collector liquid, and Stoddart et al. [63] reported an absorption system using a hydrocarbon oil as solvent.

Biogas can also contain small percentages of VOCs. These chemicals must be removed if methane is used in gas turbines and fuel cells. They can be removed by condensation, by selective membranes, by absorption and by adsorption. Silicon-based and polyether-imide based polymeric membranes can be applied.

3. Biogas Utilization

Biogas production takes place at the pressure of biodigesters, which is generally close to the atmospheric pressure. Since transport and storage require considerable costs for gas compression, biogas is generally used for the in-situ production of directly usable energy; therefore, systems for energy production must be located as close to the biogas production site as possible.

To make the kinetics of biogas production compatible with its continuous use, storage systems must be provided. Volume and pressure are determined with a compromise between investment cost (volume) and compressing operating costs. Storage should be at

low pressure and limited to the quantity apt to equalize production tips and homogenize biogas composition. Spherical gasometers are usually installed.

Energy recovery from biogas is partially used for direct self-consumption of the HSAD plant (steam and hot air production), and part of it can be used in other sections of the plant, such as heat cogeneration and production of electricity for pumps and compressors (which amounts to about 15–25% of the energy produced). The use of biogas to heat the digesters varies according to the chosen regime (thermophilic, mesophilic, etc.), the season and daily changes. Heating is generally most active when the digester is being loaded.

Excess biogas can be exploited in various ways, some of which are preferred for in-situ energy production.

An interesting overview about AD and different downstream biogas utilization can be found in Tian et al. [64]; a review on biogas development and perspective in Europe is reported in [1].

The main systems are summarized below.

Production of heat comes in the form of hot water, steam or hot air, for heating, drying and for use in other industrial processes on the same industrial site (dehydration of the digestate, purification of biogas and wastewater treatment). The average thermal efficiency is around 80–85%. The use of heating implies the existence of a local use (collective or tertiary, district heating network, industrial area).

Electricity production is used generally with gas engines, possibly with steam turbines for larger capacity plants or gas turbines. Average efficiency is around 30–35%.

Combined production of heat and electricity (cogeneration CHP). Average efficiency is about 80%, of which 45–50% is from heat and 30–35% is from electricity. As reported in [1], in high-income countries, biogas is mainly used to produce electricity and in combined heat-power plants, while heating and cooking is the main use in low-income countries. Europe is leading in electricity production from biogas.

The most commonly adopted solution is represented by cogeneration, i.e., the combined production of heat and electricity. Among the various systems used for cogeneration, which differ in the type of heat engine used for the generation of mechanical and electrical power, the following are worth mentioning: steam turbine, gas turbine, Diesel cycle engines and Otto cycle engines.

The following considerations may apply in the technical choice of the cogeneration system: the sizes of the plants most frequently installed, and the flowrate of the fed biomass and consequently of the biogas produced, which usually excludes the use of plants with steam turbines. As far as their electrical efficiency is concerned, gas turbines are on average around 10% lower than combustion engines, but seem to be a better solution in medium or medium/small capacity plants when environmental constraint are important. So far, their use has been limited by the fact that small power turbines capable of accepting large variations in fuel quantity and quality are relatively expensive. New generation gas turbines require a purity of the incoming biogas to avoid damaging blades, which is decidedly higher than that of internal combustion engines. Excellent cost/performance compromises have been obtained with small turbines, such as Ansaldo Energia's AEN-T100 [65] for applications in hotels, hospitals, laundries, farms, distilleries and breweries.

However, the economically convenient solution is in the choice of Diesel and/or Otto cycle engines. The internal combustion equipment available on the market guarantees conversion efficiencies from 30 to 42%, depending on the size of the engine and the concentration of methane in the feed gas. In order to achieve some flexibility, it is advisable to provide at least two groups, as the unit size of the motors often drops to levels where the foreseeable efficiency is between 30 and 35%. Alternator electrical efficiency is about 95%. The overall efficiency of the motor units can be considered convenient if greater than 30%; the larger the size of the plant, the greater the efficiency.

Automotive refers to the production of fuel for vehicles, especially for the supply of vehicles that transport goods. The transformation of biogas in biomethane has started to become a possible alternative to its direct use in CHP. It requires different technologies

to remove pollutants and impurities in order to be used directly as a transport fuel (by Natural Gas Vehicles (NGV)).

Fuel cells are found mainly in the automotive sector. The first studies and demonstration plants started by using stationary phosphoric acid fuel cells (PAFC). A 200 KW pilot-plant, operated with cleaned biogas produced from AD, is described in [66].

The most used are PEMFCs [67], for which integrated reformers are usually adopted. The hydrogen produced is purified downstream with conventional processes or through water gas shift [68].

Some studies have also been conducted for the use of biogas in molten carbonate fuel cells (MCFCs), like [69,70]. The first European MCFCs using biogas from AD is described in [71]. MCFCs with integrated reformer (MSR), which has typical requirements of less than 0.05 ppmv of sulfur, halogen and silicon compounds, requires strong biogas treatment, as discussed in [72], since they cause irreversible poisoning of the Ni-based catalyst of the MSR [73].

Finally, some applications also exist with Solid Oxide Fuel Cells (SOFCs) using biogas produced in wastewater treatment plants [74], anaerobic digestion of waste [75] or organic waste [76,77]. A recent review on SOFCs fueled with biogas can be found in [78].

Currently, the practical utilization of upgraded biogas in fuel cells at industrial scale has minimal relevance, mainly due to economical reasons.

Other possible applications include the production of natural gas for injection in public transport and distribution networks and the production of cold; for example, absorption machines are useful in agro-food industries that feed digesters with the waste biomass they internally produce and also need to preserve food.

Finally, another application involves the use of biogas in industrial ovens as primary or auxiliary fuel [79].

4. Operating Modes, Reactors and Stages

The scientific literature concerning the comparison of plants layouts, type of reactors and operating conditions is extensive. Given the complexity of the multiphase system, in which chemical and biological reaction networks involving different types of microorganisms take place, many design and operational choices are possible. This includes the type of reactor and the ranges of the main process parameters, but also the possibility of separating the microorganisms involved in the different phases of the anaerobic digestion (hydrolysis, acidogenesis, acetogenesis, methanogenesis), i.e., operating in double-stage, with each stage set at different process conditions. From an industrial point of view, the objective functions mainly used to guide these choices are the quantity of biogas produced and/or the concentration of methane in the outgoing gas (to be maximized), energy costs, land use, water resources (both to be minimized) and investment costs (to be minimized).

The most important choices regarding AD processes can be summarized as: (1) the operating mode, (2) the type of reactor and (3) the number of stages (see Figure 1). These choices affect CAPEX, land use and performance of the control strategy.

The second group of conditions on which to make a choice regard the two main process parameters: temperature (mesophilic or thermophilic conditions [80–82]) and total solids in input [8,83]. They can affect operating costs and maintenance costs, as shown in Tables 2 and 3.

Once the optimal choices have been made according to the objective functions appropriate for each application (which also depend on the type of biomass), all the optimization and improvement margins depend on how much we are willing to invest in automation and process control.

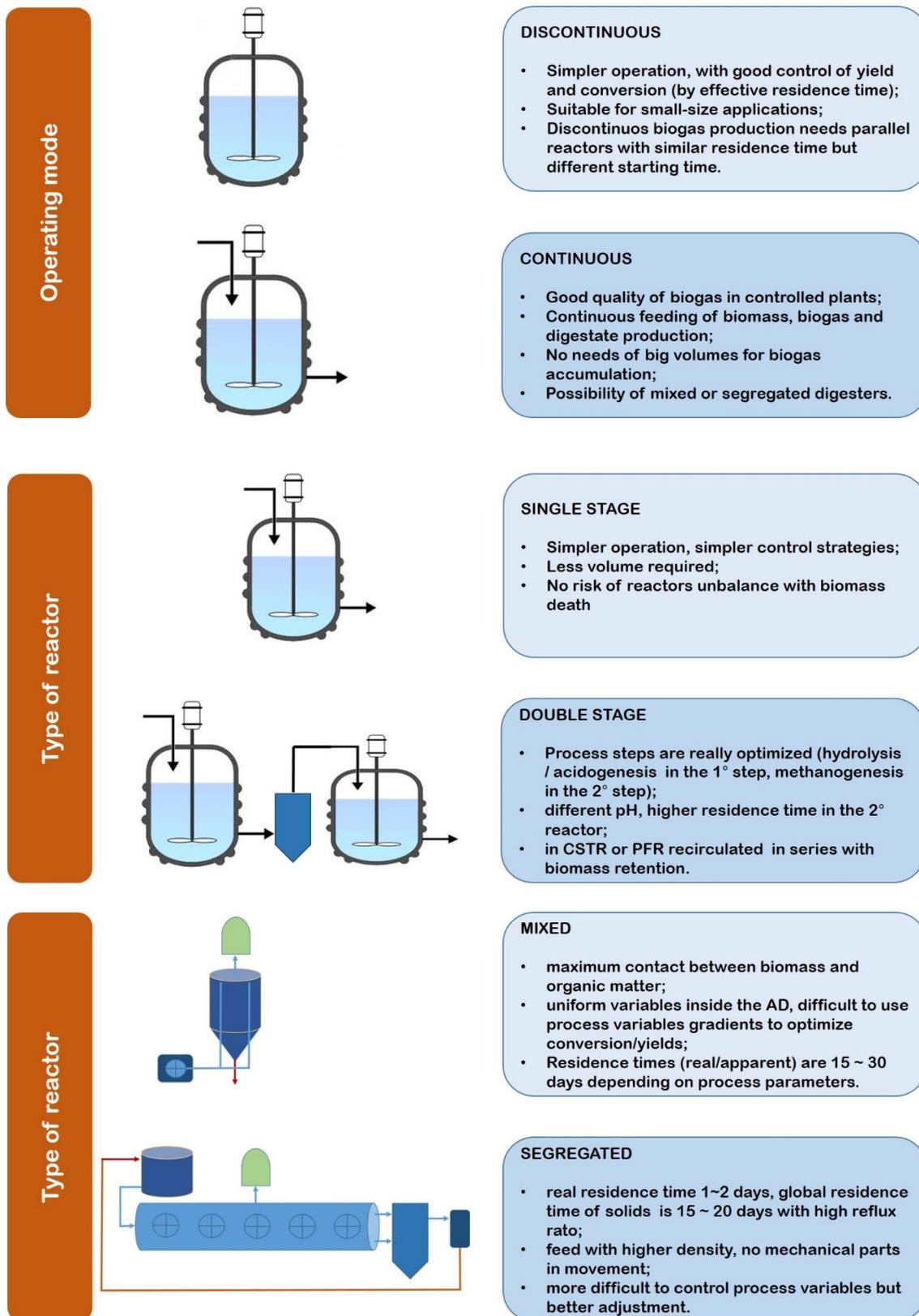


Figure 1. Main choices about operating mode, reactors and number of stages.

Table 2. Thermophilic vs. mesophilic conditions.

Operating Parameter: Temperature	
Thermophilic 55 ± 2 °C	Mesophilic 35 ± 2 °C
higher yields in biogas smaller volumes higher energy consumption enhanced diffusion of organic substrates to the microbial cells	higher hydraulic retention time (HRT) better diversity of methanogenic microorganisms higher volatile solid reduction

Table 3. High Solids vs. Low Solids content.

Operating Parameter: Total Solids	
HSADs Up to 50%	LSAD Up to 15%
does not need waste pre-treatment (dimension reduction to about 4 cm) usually uses segregated reactors (no mixing requested) lower residence times lower costs (volumes, pumping and energy)	more consolidated needs dilution with water mixing problems are often observed, with by-pass or separation higher volume possibility to correct process parameters and peak values by dilution simpler pumping systems (liquid biomass)

5. Data Driven Models

5.1. Phenomenological Models

Phenomenological models do not consider a priori information about the process, and their characteristic variables are identified during the model construction. At first, the parameters inside these models have no physical meaning and are those for which maximum compliance with the historical series of available data are obtained. Experimental data (time series for dynamic models) are also used to determine the shape of the model within a class of functions, suitable for linking the identified variables in cause-effect relationships. This means that both parameters and order of polynomials are estimated from data [84]. The term phenomenological model is often used as a synonym for black-box models; however, the class of phenomenological models can also include empirical models by describing what happens inside the system under study. Therefore, if the phenomenological model uses the state variables in the empirical relationship, it is a white-box (or grey-box) model. If the model equations only link the input and output variables, the model is a black-box type.

The difficulty in developing reliable physically-based mathematical models for describing HSADs' dynamics [85] is due to the non-linear and complex dynamics of biochemical processes, which require detailed knowledge on physical, chemical and biological mechanisms at different scales inside the reactor and in living cells. This includes the use of many variables and the characterization of a high number of parameters to be inserted into the model. This high number of variables and parameters affects computational complexity and sometimes influences the model identification procedure, requiring a "rich" time series of all the considered variables in order to correctly estimate the high number of model parameters. Conversely, the structure of phenomenological data-driven models is relatively simple; therefore, it could be used efficiently for simulation of HSADs since only the interesting variables are introduced into the model and risk of parameters overestimation is reduced.

The phenomenological approach to HSADs modeling has increased in the last twenty years, due to the availability of fast algorithms for data filtering and management for model identification and machine learning. Approaches can be grouped in classical black-box

models or in Neural Networks (NN) and machine learning techniques, which are sometimes considered grey-box models, if new physically based correlations are individuated after the learning process.

Phenomenological models can be identified using less “rich” time series than those required to identify physically based models, and data can be acquired during start-up, shut-down and normal operation of the existing plants. In order to construct robust dynamic models it is important to design proper experimental runs. Acquiring data during operation means to previously define admissible and acceptable ranges of each input variable; subsequently, the plant should be operated by changing inputs inside these ranges and according to disturbance series (steps, pseudo random binary, pseudo-random, white noise, etc). The frequency of variation depends on the characteristic time of the considered input variable. The outputs of the plant are captured during transients. Situations that lead to unacceptable values of the output variables (yield, biogas composition) can be useful to construct the model. The holdout method is usually adopted, by selecting half data for model identification (training) and the remaining data for model validation.

5.2. Black-Box Models

Time series based black-box models for HSADs can be generally structured as multiple-input multiple-output (MIMO) models, whereas good results can be obtained by considering a MISO scheme, where methane flowrate or yield is the only considered output variable.

By grouping the measurable variables of the system in input $\mathbf{u}(t)$ and output $\mathbf{y}(t)$ variables, a general non-linear input-output model can be written as $\mathbf{y}(t) = \mathbf{f}(\boldsymbol{\psi}(t), \mathbf{p})$, where $\boldsymbol{\psi}(t)$ is called the regression vector and \mathbf{p} are the specific parameters of the black-box model. The identification procedure consists of using the acquired dataset to define the shape of the function and to estimate the parameters. The regression vector contains the input variables, the previous outputs and errors. The function f can be expressed as:

$$f(\boldsymbol{\psi}(t), \mathbf{p}) = \sum_{k=1}^n a_k f_k(\boldsymbol{\psi}(t), \mathbf{p}) \quad (1)$$

where $f_k = \sigma(\beta_k(\psi - \gamma_k))$ and σ is a mother basis function, β and γ are, respectively, called dilation term and translation parameter [86]; n is the number of terms required to acquire a certain degree of approximation (details can be found in [87]). The choice of these permits approximate f_k as polynomials, Fourier, sigmoid, wavelet functions.

Linear black-box models can be summarized by the general family [88]:

$$A(z)y(t) = \frac{B(z)}{F(z)}u(t-n) + \frac{C(z)}{D(z)}e(t) \quad (2)$$

where z is the shift operator, so $A(z)$ is a polynomial in z^{-1} .

When both $F(z) = 1$ and $D(z) = 1$, Equation (2) reduces to the linear AutoRegressive Moving-Average with exogenous term ARMAX model written as:

$$A(z)y(t) = B(z)u(t-n) + C(z)e(t) \quad (3)$$

Special derived cases from Equation (3) are known as ARX model when $C(z) = 1$, ARMA model when $B(z) = 1$, AR model when both $B(z) = 1$ and $C(z) = 1$ and finite impulse response model FIR when both $A(z) = 1$ and $C(z) = 1$.

When complex non-linear dynamics modeling is requested, the corresponding NARMAX models can be adopted. They can describe complex nonlinear dynamics, as multiplicity of steady states [36], oscillation between thermophilic and mesophilic conditions, shut-down of the digester due to biomass variation. Although these models are robust and reliable, there are very few applications on HSADs, since the scientific community prefers to devote efforts to possible applications of Artificial Neural Networks (ANNs). A comparison between two ARX models (SISO and MIMO) with a same order of ANNs' model on a fluidized bed anaerobic digester was proposed by Premier et al. [89].

A greater number of applications of black-box models are related to wastewater treatment processes, which in some ways share a large part of biological mechanisms with AD processes. Among them, Novotny et al. [90] proposed an ARMA model for describing the effects of BOD and SS on MLSS concentration; Capodaglio et al. [91] proposed both SISO and MIMO ARMAX models; Van Dongen and Geuens [92] and Sotomayor and Garcia [93] proposed derived ARMAX models to evaluate MLS and TSS from BOD, COD, SVI data.

Despite the current preference of the scientific community for ANNs and machine learning, time series based black-box models remain, in our opinion, a very promising approach, given the recent studies searching for cause-effect relationships between input and output HSADs variables. In many of these studies, methane yield or methane quality is considered the output variable and the biomass composition the main input variable, so the applicability of these models is relatively simple. Algapani et al. [94] correlated food waste and sewage sludge composition to methane production and Vivekanand et al. [95] and Valenti et al. [96] investigated the relationships between different co-digesting biomass and methane yield. Jin et al. [97] correlated how corn straw and pig manure co-digestion influence HSADs performance; López González et al. [98] defined the effects of sugarcane input on methane production; Marques et al. [99] discussed co-digestion of biorefinery wastes; Thorin et al. [100] investigated the performance of co-digestion of sewage sludge and microalgae.

5.3. Machine Learning

The term Machine Learning (ML) groups all the actual algorithms that permit construction of phenomenological predicting models starting from data. These models can predict outputs of complex nonlinear processes like HSADs. In this sense, the produced models are black-box type. By combining ML with data mining, some new properties or new constitutive equations can be discovered for some state variables; in this case, grey-box models can be produced. In order to construct models based on ML, the type of learning and the network architecture must be defined and a large dataset containing all the possible measured variables of the process must be provided. Differently from classical black-box models, it is not strictly necessary to previously group variables in input-output groups. The learning techniques adopted in artificial neural networks (ANNs) have many similarities with the parameter estimation algorithms used to find both parameters and model in NARMAX black-box identification schemes.

Many studies can be found in literature regarding the application of ML to lab-scale HSADs processes, and in most of the applications ANNs are used and methane yield or its flowrate is still the only predicted output variable. Examples can be found in Mahanty et al. [101] where an ANN was used to predict methane production with different types of biomass, Jacob and Banerjee [102] used an ANN combined with genetic algorithms (GA) to predict and optimize methane production from input potato waste and aquatic weed and in Xu et al. [103] ANN is used to predict methane yield in a mesophilic SSAD.

Different output variables have been considered in Sinha et al. [104], where ANN was used to predict the performance of a UASB, in terms of average gas production rate and methane percentage in the biogas used as input in the organic load flowrate, the hydraulic retention time, and inlet bicarbonate alkalinity. The ANN was also used to predict the effluent substrate concentration and some state variables, like bicarbonate alkalinity, pH, volatile fatty acid concentration in the reactor. Wang et al. [105] applied ANNs to predict alkalinity in an anaerobic co-digestion process by using pH, ORP and EC data and Antwi et al. [106] used a feed-forward back propagation ANN to predict COD removal in a UASB reactor treating industrial starch processing wastewater.

ANNs are often combined with neuro-fuzzy inference systems like ANFIS platform, and are able to treat qualitative data as discussed in Pai et al. [107] to predict COD and SS in a WW treatment plant. De Clercq et al. [108] demonstrated the strong predictive power of ML Algorithm XGBoost applied to biomethane production modeling, based on industrial-scale ACoD project data.

6. Process Control

Dry ADs are generally more complex than wet AD systems in terms of process conditions, particularly due to two reasons: the higher organic loading rate and the lower hydraulic retention time increase input sensitivity and reduce the characteristic response times of dry ADs; the acceptable ranges of the process variables are usually narrower in dry ADs. The complexity, from the plant engineering point of view, is also greater in these systems, such as in the AnMBR, in the up-flow anaerobic sludge blanket (UASB) reactors or in the expanded granular sludge blanket (EGSB). For these reasons, although many of the control strategies used for traditional AD plants can be extrapolated for these plants, it is usually preferred to adopt more sophisticated technologies for HSADs processes. Furthermore, investments in the adoption of advanced control systems are justified for HSADs due to the relatively high productivity, and consequently high performance of these plants, if compared with traditional AD with the same volume.

The reasons that lead to further research and new design of reliable and robust control strategies for these systems, can be briefly summarized as follows:

- (1) Environmental legislation and safety;
- (2) High noise on the input variables, most of which cannot be manipulated;
- (3) High number of state variables influenced by the inputs;
- (4) Low number of controllable output variables.

Leaving aside the control problems during the start-up, which need a separate discussion, the amplitude of disturbances on the input variables is high, even during normal operating conditions, and the frequency of the disturbances is hardly predictable. The greatest fluctuations occur on the flow rate and on the composition of the influent to be treated, which translates into variations in COD, TSS, FOG and on the possible presence of toxic substances not expected under normal operating conditions. Inlet temperature and pH can also be subject to disturbances. The frequency of the disturbances can also be high, if compared with the apparent residence times inside the reactors, which correspond to variations of a few hours.

Among the input variables, many of them (although in theory all measurable even if at high costs) cannot be manipulated, as concentrations in the input flows. Instead, the input flow can be manipulated in terms of recirculation of liquids/sludge at the input. Acidity can also be manipulated by adding bases. The dilution rate is the easiest input variable to manipulate, together with some strictly plant dependent variables such as agitation and mixing (by acting on the biogas recirculation flowrate or steam insufflation in thermophilic processes).

Internal variables, or direct effects on them, are overload, acidification, foaming, inhibition, and lack of macronutrients and trace elements.

The main output variables are biogas flowrate, which is obtainable by direct or inferential measurements (i.e., measurements of VFA and VFA/TA as in Zhou et al. [109], COD and VFA in the effluent [110,111], dissolved hydrogen, pH and alkalinity. Among them, the biogas flowrate is usually the main controlled variable.

In co-management systems one substrate is usually the controlled variable and control action is exerted by manipulation of dilution. Membrane fouling can be the controlled variable in AnMBR by manipulating washing water, as shown in Smith et al. [112], and gas sparging, as shown in Park et al. [113]. HRT and SRT can be controlled as proposed by Robles et al. [114].

The HSADs' process dynamics is highly nonlinear, and this means that closed loops become nonlinear if linear controllers are adopted. Different combined control strategies have been proposed during the last fifty years. Some reviews can be found in Nguyen et al. [115], Jimenez et al. [34] and Gaida et al. [116]. In addition to the classic controllers, fuzzy control and neural networks have been proposed, such as model-based control strategies, adaptive control and feedforward control loops.

A brief summary of the main adopted control methods is reported.

6.1. Open-Loop and On/Off

The manipulated variable is set to a binary or multiple-value, depending on predefined threshold values. This type of strategy cannot take into account the high variations of the input flowrate, organic load and pollutants. It was the first control strategy proposed about fifty years ago. This control strategy is not used and can only be adopted in very small units.

6.2. Closed-Loop with PID Controllers

PID controllers, including P and PI are classical, relatively low-cost, closed-loop systems. These regulators can be adopted to stabilize pH by manipulating the introduction of chemicals or to control the biogas flowrate, as shown in von Sachs et al. [117]. Standard PID are SISO systems, so PID cascades are usually proposed with two distinct set-points, one on the biogas flowrate and the other on the organic load, which are combined with different control strategies, as shown in Alferes and Irizar [118]. Servo-controllers can be designed, adapting the biogas flowrate set-point to the behavior of a specified process variable, as shown in Zhou et al. [60], where the ratio VFA/TA is used, or in García-Diéguez et al. [119], where the effluent VFA is the specified process variable. Also programmed-value PID systems allowing modification of the set-point during plant operation are possible options.

6.3. Adaptive Controllers and Adaptive Control

Due to the high non-linearity of the process, PID controllers or PID cascades must adapt their control gains depending on some process variables and/or plant state. The control gains of the recently adopted PID controllers are at least modified, from start-up conditions to continuous operation of HSADs.

Among the classic adaptive control strategies, linearization is the most used. Linearizing control strategies were used in Ignatova et al. [120] and Méndez-Acosta et al. [121]. This kind of control strategy works well only in a definite linearized zone in the neighborhood of the set-point, due to the high nonlinearity of the process dynamics. Moreover, this kind of strategy requires model identification, which needs high computational efforts so often that surrogate models as black-box are proposed. In this case, the model identification procedure allows selection of the best model among similar classes. It is to be noted that linear black-box models, such as ARMAX or ARMA, can be adopted, since they can describe nonlinear dynamics. NARX models, which require simple model identification algorithms as least squares, can work well since they can also describe complex dynamics like multiplicity of steady states [36].

State-space models can be used, or input-output linearization. Finally, interval-based approaches, similar to linearizing control, can be adopted to stabilize the regulated variable in a neighborhood of the set-point, as proposed in [122].

Interval observers-based adaptive control uses the values of some variables called observers (related to the chemical-physical relations between some internal variables) to regulate the controlled variable on the set-point, as shown in Aguilar-Garnica et al. [123]. Finally, sliding control is also used due to its robustness, as shown in Lara-Cisneros et al. [124].

6.4. Expert Systems

AD was among the first processes in the 80s where expert systems for process control were tested. This is mainly due to the higher response times of the entire process (i.e., relative slowness of the transients) then those of electrical processes, energy production or chemical/pharmaceutical processes with exothermic reactions, that are at runaway risk.

Previously, expert systems were classified into rule systems (logic/deterministic or fuzzy logic based) and systems that use surrogate models (based on neural networks, for example). Nowadays, these systems use all the possible combinations of different methodologies to implement their knowledge base.

Some early applications of expert systems considered monitoring, supervision and control of wastewater treatment and AD plants [125–127]. Examples of the application of fuzzy

logic as a control strategy can be found in Estaben et al. [128], Pullammanappallil et al. [129], Murnleitner et al. [130] and Carlos-Hernandez et al. [131].

A mixed approach using an expert system based on fuzzy logic coupled with a classical PID controller was proposed by Heredia-Molinero et al. [132] to control pH. A combination of neural networks and fuzzy logic can be found in Tay and Zhang [133] and Waewsak et al. [134], where the neural network is used to predict some output variables in advance, such as biogas flowrate, VFA and TOC, fuzzy logic is also adopted to manipulate the input variables, like influent flowrate.

6.5. Other Control Schemes

A linear mixed feedforward/feedback control scheme was proposed in Méndez-Acosta et al. [135] in order to reduce load disturbances due to the noise in the inlet composition.

Liu et al. [136] developed a cascade controller system that is embedded into a rule-based supervisory system based on Extremum-Seeking-Control (ESC), where the operating set-point is sought by optimizing a determined performance function. Alferes and Irizar [118] used ESC combined with a fuzzy based supervisory module to optimize biogas production and modify the AD plant set-points; Lara-Cisneros et al. [124] proposed a combination of ESC with sliding mode to optimize biogas production. Superimposing a probing signal to the feed flowrate was proposed by Steyer et al. [137] in order to collect information on the plant output (biogas yield) and to determine the operating set-points, and consequently, decide if the influent flowrate should be increased or decreased.

7. Discussion and Conclusions

Since the industrial sector analyzed in this work allows for the production of biogas from wastewater and organic waste (therefore from a starting material considered a waste), it is classified as a sector requiring a “low level of automation”. Although it may have been true previously, automation investment costs may not be sustainable when compared to the value of the product (dirty biogas and 50% methane). The current significant reduction in fossil energy sources, the need for decarbonisation and the recent geopolitical situation lead to reconsiderations around economic evaluations regarding the advantages obtained in the control of these plants. This is evident in light of the rapid reduction in hardware and software costs and the possible adoption of open hw/sw.

There is high research activity in data-driven models proposing black-box input-output NARMAX models and ANNs, but the results obtained at laboratory scale or in small pilot plants are rarely extended to large production plants. For example, the multiple linear regression model proposed by Rossi et al. [138] predicts the specific methane production from a pilot plug-flow dry anaerobic digester, which could be implemented at industrial scale to improve production from existing plants that treat the organic fraction of MSW.

A similar example includes the methodologies and control strategies proposed by the academic community, which are rarely adopted at industrial scale.

Moreover, the biogas produced with the recent High Solids Anaerobic Digesters can reach a methane concentration of about 65%; however, several treatments are still required, including desulphurization and the removal of siloxanes. In addition, for some applications like direct domestic use of bio-methane or gas turbine power generation, the treatment of potential pollutants needs to be more stringent. The strategies for indirectly reducing biogas treatment costs are undoubtedly to be found in better understanding the process, the construction of dynamic predicting models and process optimization and control.

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