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A call for modernisation of science, the case of anaerobic digestion: A scoping review

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A B S T R A C T

Anaerobic digestion is an important renewable energy technology that has the potential to reduce greenhouse gas emissions and contribute to the development of a sustainable energy system. However, research on anaerobic digestion is extensive and fragmented, making it difficult to gain a comprehensive understanding of the technology. Therefore, a large scoping review was performed, using adapted PRISMA protocol, aims to provide a comprehensive overview of the anaerobic digestion process, from pretreatment to gas utilisation, and the research conducted in each step. In addition, this research serve to highlighted issues in the current research environment as to identify limitations. The review involved analysing 4660 articles and exhaustively identifying the different methods for following research parameters: pretreatment (1279), reactor design (923), temperature (907), H_2S cleaning (893), and biogas upgrading (658). This information can serve as a basis for future standardisation of work to increase the efficiency of biogas research.

There is a large number of methods employed in all the parameters explored, with a lack of standardisation in reporting and no clear definition for certain terms leading to confusion and issues classifying. A problem exacerbated by an exponential growth in published research. This also compromises comparisons and reviews accuracy and leads to tedious work to recover information and the inevitability of missing crucial information. As a result, we observed a lack of exhaustiveness in this work. This also highlights the urgent need for harmonisation of research to facilitate knowledge transfer and avoid redundant work or the overlooking of potential breakthrough research, while also indicating to researchers where focus should be orientated. Modern databases and centralisation could help bring standards, simplified reviews and updatable work hampered by traditional reporting. Research needs to be more accessible and rigorous using the open science framework, as to promote transparency, reproducibility and cooperation.

1. Introduction

Scientific efficiency is vital to address the urgent challenge that is climate change. Bioenergy, constituting a substantial share of renewable energy sources in the EU, stands out as a crucial player in mitigating global warming[109]. Biogas, which is produced through the process of anaerobic digestion where organic material is decomposed by a microbial consortium in the absence of oxygen, is a major technology in this category and plays a central role in some EU member policies [45]. This technology, not only serve multiple energy purposes, such as heating, electric generation and grid integration through methane upgrading. It is also a valuable waste management tool, helping reduce waste and avoid harmful emissions[43]. It has, for example, a large potential in food waste management as industrial sector waste equals 5 % of its total food production, and this waste is underutilized[97].

The current scientific landscape has seen an exponential growth in publication since its inception in the 17th century[17]. This is present as the same time as the mention of a 'reproducibility crisis' in the science community, with more than 70 % of the researchers failing to reproduce an experiment from another scientist[10,30]. Anaerobic digestion research is probably not immune from this phenomenon and would need some careful analysis to uncover the state of the field and the steps necessary to ensure its efficiency for the technology to fully play its role. To alleviate these issues with science, there has been a push towards 'open science'. The concept does not bear a precise definition or unified view but is a new vision for conducting science with more transparency. An

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attempt to form a rigorous definition would be the following: 'Open science is transparent and accessible knowledge that is shared and developed through collaborative networks' [126]. There have been attempts to provide guidelines to perform more open science encompassing: open access; open material, data and code; reproducible analyses; preregistration; replication research; and teaching open science [30]. One noteworthy initiative exemplifying this shift is the PRISMA (preferred reporting Items for systematic Reviews and Meta-analyses) 2020 protocol. It is a reporting guideline first published in 2009 to cover a need in health science, where poor reporting standards undermined the critical role of systematic reviews in the synthesis of data to draw meaningful conclusions. It quickly outgrow its domain to encompass all area of science and was thus updated through time with the latest statement published in 2020. It provides a checklist of items to follow, to ensure transparency and reproducibility in review articles[91].

To be able to push the field further, a good global vision is necessary in the first place. As in publications in general, the numbers of articles published in this field is likely increasing too and would require a good organization. Currently, most review articles are not systematic, meaning that they do not disclose how they collected their source and reflect more the author knowledge of the field. This does not mean that articles cannot seem exhaustive and can reflect a good portion of the technology of a particular field. For example, looking for reactors design in wastewater would give good source, but without any assurance on exhaustiveness[120]. Articles covering actual industrial applications, which is a bridge often missing in science also exist, albeit in this case on a reduce sample of a few company designs[55]. Age is an issue and new breakthrough techniques might be missed in older sources. For emerging technologies specifically, reviews exist covering specific areas, such as for pretreatment[96], but such work are not necessarily updated regularly and newer reviews usually focus on different subjects[106], due to pressure on publication, impact and novelty demanded[46]. Moreover, the lack of methodology raises a question on the exhaustiveness of such works. The lack of details on the specific methods with a focus on more general classifications, can be an issue for certain researchers trying to innovate with new methods, especially with no methodology to be able to reproduce those results. Systematic reviews should add the clarity and order, they might not be perfect as they do not allow for article selection based on quality. Recently more articles with a systematic approach were published, albeit the methodology description might be still quite lacking, [88], or missing crucial features such as the description of exclusion and inclusion criteria[54]. Overall the issue of details still remains in systematic reviews, where an article covering biogas usage classify the different methods without going into the details of all the specific subjects [79], and might miss certain features like fuel cell in this case[41], not necessarily by the authors fault as systematic review does not allow for the choice of articles. Unfortunately, systematic reviews do not mention difficulties at performing the work and what adaptation would be necessary as to allow for faster and more accurate work. The current review landscape suffer from information fragmentation, where it is difficult to find all relevant information in one place and ensure the discovery of relevant information. It also remains to be seen if performing such an exhaustive work would be possible.

There are multiple steps to be taken to produce biogas. Currently, there is a wide range of research covering all these steps of anaerobic digestion. Unfortunately, the research on anaerobic digestion is extensive and fragmented, with little standardisation, making it difficult to gain a comprehensive understanding of the technology. Consequently, the gathering of data and performing a quality summary of the field can be complex. An exhaustive review covering the entire topic is needed. Currently, it is hard to find specific information or new techniques with potential. It also leads to a struggle for new researchers in the field to grasp the complexity of the field and reduce their ability to perform their work efficiently.

This work aims as first goal to attempt at giving some order to this situation by performing a broad exhaustive scoping review, systematically listing the evolution of research in the field and the areas of scientific focus. This should help new researchers understand the field better and future research to understand where there is a potential research gap or guide the development of a new digester type in the areas of focus. This research does not aim to explain the benefits or drawbacks of all the methods listed. This should be covered in specific Systematic reviews. However, this review can help indicate where such a review would be needed. The secondary goal of this research is to highlight the issues encountered in such a work, using lessons learned from open science initiatives to better reproducibility and transparency. This should give us a clear picture of the situation in anaerobic digestion and give us insight to the shortcomings of the current research environment and ways to improve it.

2. Methods

The PRISMA (preferred reporting Items for systematic Reviews and Meta-analyses) 2020 protocol is a reporting guideline first published in 2009 to cover a need in health science, where poor reporting standards undermined the critical role of systematic reviews in the synthesis of data to draw meaningful conclusions. It quickly outgrow its domain to encompass all area of science and was thus updated through time with the latest statement published in 2020. It provides a checklist of items to follow, to ensure transparency and reproducibility [91]. It was adapted to use in this context and thus only the first 9 items were followed.

This review aimed to identify the existing processes for 5 of the major parts of biogas production, namely pretreatment, reactor design, temperature, desulphurization, and biogas upgrading. These parameters were chosen because they are design choices that must be planned and crucial to the operation of any anaerobic digestion process. The articles would be reviewed according to the following process: article database create through Web of science, screening to only include relevant articles and then divide the articles into 2 parts (research articles and reviews) for further classification (Fig. 1).

Web of Science was used to identify eligible studies, the research review article from 1945 to 2022. The research is based on the authors' keywords. Unfortunately, there is no standard way of reporting keywords in this field; hence a combination of different ones was used to ensure a larger, more exhaustive sample set. The following prompt was used in Web of Science to provide results about anaerobic digestion '*AK=(biogas OR "anaerobic digest*" OR biodig* OR biomethan*)*'. The first keyword (biogas) refers to the main output of the technology. Second, anaerobic digest* was included because it can refer to the underlying biological process of anaerobic digestion or, less commonly,

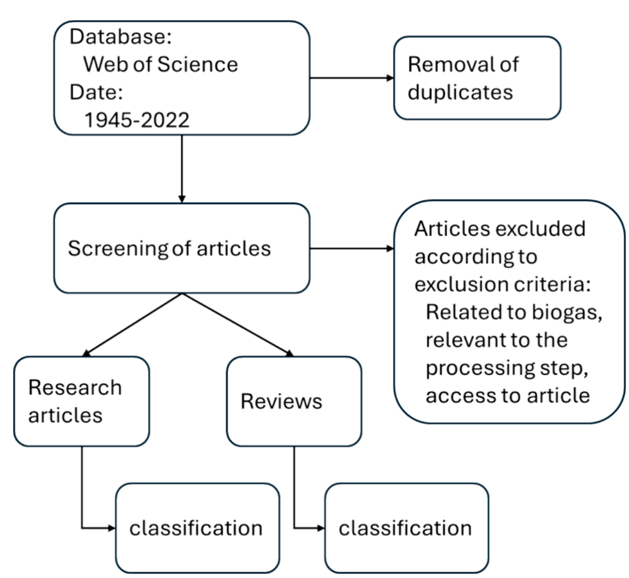

Fig. 1. Flow diagram of the methods used for articles screening.

anaerobic digester as a processing unit. Biodig* is a keyword that can mainly refer to biodigestion or biodigester, two common names for the process and the processing unit. Lastly, biomethan* (biomethane/biomethanization,…) refers to the gas of interest, namely methane obtained through the process of anaerobic digestion.

This combination of the previous prompt and one more specific to each processing step were then used to provide the complete database:

- Pretreatment: '*((AK=(biogas OR "anaerobic digest*" OR biodig* OR biomethan*)) AND AK=(pretreat*))*'. This keyword can refer to: pretreatment, pretreated, pretreating etc.
- Temperature: '*(AK=(Biogas OR "anaerobic Digestion" OR biodig* OR biomethan*)) AND AK=(psychrophilic OR mesophilic OR thermophilic OR hyperthermophilic OR hyperthermophilic OR hyper-thermophilic)*'. For the temperature, authors generally do not refer to the temperature in keywords but refer to the temperature range employed directly. The keyword arrangement had to be adapted to include all possible temperature ranges.
- Reactor: '*(AK=(Biogas OR "anaerobic Digestion" OR biodig* OR biomethan*)) AND AK=(reactor)*'. For the reactor design, there is no perfect solution as the authors often refer to the design, they used ex: UASB or CSTR instead of mentioning the keyword 'reactor'. Such articles would be missed, but there is no keyword that could provide a better solution to encompass all reactor designs.
- H_2S: '*((AK=(anaerobic digestion OR biogas OR biodig* OR biomethan*))) AND ALL=(H2S OR "hydrogen sulfide" OR "hydrogen sulphide" OR desulphurization OR desulphurization)*'. This combination of keywords refers to the gas of interest in its different nomenclatures and to the removal process.
- Biogas upgrading: '*(AK=(biogas OR "anaerobic Digestion" OR biodig* OR biomethane*)) AND AK=(upgrading)*'. No other keyword was deemed necessary, as it is standard to call upgrading from biogas to biomethane in such a way.

For each step, the resulting article list was exported to excel for further review, with the following parameters: title, keywords, abstract, year of publication, and source. One researcher oversaw reading the abstracts of all imported articles were screened by the main author, and a study was excluded if the objective was not to produce biogas through anaerobic digestion. For this reason, cases such as hydrogen production through anaerobic digestion, and biogas obtained through other means were excluded. Secondly, an article needs to mention the use of the processing step. In case the actual process cannot be identified, the article will be reviewed directly for more information. If it is still not clearly stated or access, through the Czech university of life science, to the article is not possible, the article was excluded. Due to the large scope of the research, it is not feasible to request the article from the authors if clarifications are necessary.

The actual collection of data records if it is a review article or else specific information according to the specific part. For the pretreatment part, the method used was recorded along with the substrate and if it was used for solid digestion. For the reactor design, the design, size of the reactor in 3 categories (laboratory scale, pilot small scale or full scale), where we included in pilot scale all reactors going from 100 L to a couple of cubic meters. the substrate, and if it was used for solid digestion. The temperature range, the order (for when different temperatures are used in multiple-stage reactors), the substrate, and whether it was used for solid digestion were recorded. For hydrogen sulphide only, the method used was recorded, and for biogas upgrading, the method used and if it was used in situ of the reactor or ex situ. Afterwards, according to the results obtained, the data would be divided into categories, based on common definitions, and plotted over the years to observe evolutions. Lastly, the reviews would be classified according to these categories.

3. Pretreatment

Pretreatment is often necessary, as not all substrates can be used directly in anaerobic digestion. Some cannot be digested by microorganism, or would be too slow, thus impractical, while others contain inhibitors that can reduce microbial activity [96]. There are different strategies to increase bioavailability, reduce hydraulic retention time, and increase biogas production of recalcitrant material, with direct degradation of the structure or increased surface area for enzymatic degradation. Lignocellulose biomass is abundant, but underutilised due to its recalcitrant nature. It is composed of lignin, hemicellulose, and cellulose. Different classifications exist for its pretreatment: physical, chemical, biological, and physicochemical is one possible way used for lignocellulose [1]. Algae are also a challenging material to digest due to their complex polysaccharides. The classification used in this case divides the methods into categories: physical, chemical, biological and thermal [122]. Sewage sludge contains complex structures, recalcitrant cell walls, and potential pollutants that need to be managed. Some strategies are classified as mechanical, biological, thermal, and biological [57]. Unfortunately, with no coherent classification method. Reviews will include methods and classify them according to the authors' knowledge and lack systematic methodology.

For the pretreatment, 1279 articles were found from 1991 to 2022 (see Fig. 2). Of these articles, 84 were rejected according to the exclusion

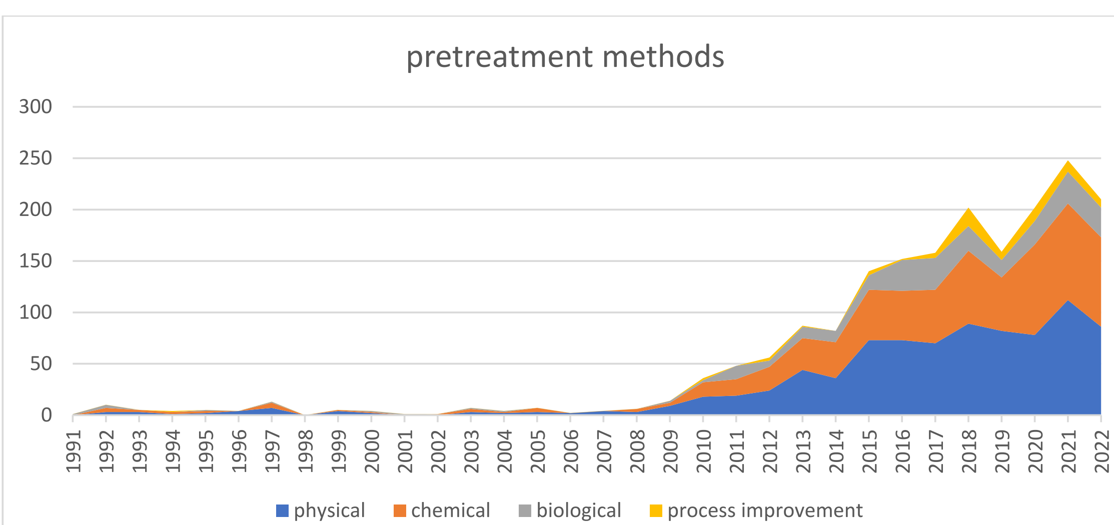

Fig. 2. Count of all the methods used for pretreatment divided into 4 groups: physical, chemical, biological, process improvement, from 1991 to 2022.

criteria, some articles talked about ethanol production or hydrogen production, while in some cases anaerobic digestion was considered as the pretreatment itself for some substrates. Unfortunately, multiple articles had to be excluded due to a lack of accessibility to the article. Another 140 articles were reviews.

The methods obtained were divided into 4 groups as shown in Fig. 2. The largest amount of pretreatment found was of physical nature, with 856 references to a physical method in scientific articles. The next group was chemical with 692 references followed by biological methods with 254 references and process improvement was less numerous with 75 references. Each group was then further divided in different categories as seen in Fig. 3.

3.1. Physical methods

Physical methods can be defined as methods that do not require additional compounds to increase the bioavailability of the substrate [6,104]. This means in practise the use of energy to break down material. Several methods can be used, as shown in Fig. 4. The most prevalent methods are of thermal nature with 563 references. Ultrasonic and mechanical disintegration methods are second with 123 and 109 references respectively. Pressure (24), electrolysing (19) and freezing (13) methods are more anecdotal. Lastly pulsed electric field is a new technique with only 5 references.

Thermal methods includes many heating methods. The methods observed were microwave, hydrothermal, steam explosion, autoclave, thermal hydrolysis, liquid hot water, thermal pressure, steam, torrefaction, wet explosion, cooking, hot air, pyrolysis, saponification, steam cracking steam distillation, boiling, as well as references to heating at low temperature.

Ultrasound increases bioavailability through the production of cavitation that induces free radicals and pyrolysis inside the caveating bubbles [49]. Furthermore, ultrasound generates heat inside the substrate that can help to degrade it, albeit at low efficiency [142].

Mechanical disintegration methods were found under these terms: grinding, milling, extruding, shearing, and chopping.

Pressure-based methods (24) usually revolve around a sudden change in pressure. Hydrodynamic cavitation also produces cavitation but at a more efficient rate than ultrasound [142]. Other mentioned methods are high pressure homogenisation, shock treatment and mechanical jet. Some methods that use pressure are not considered here, as they use heating with an increase of pressure due to an enclosed

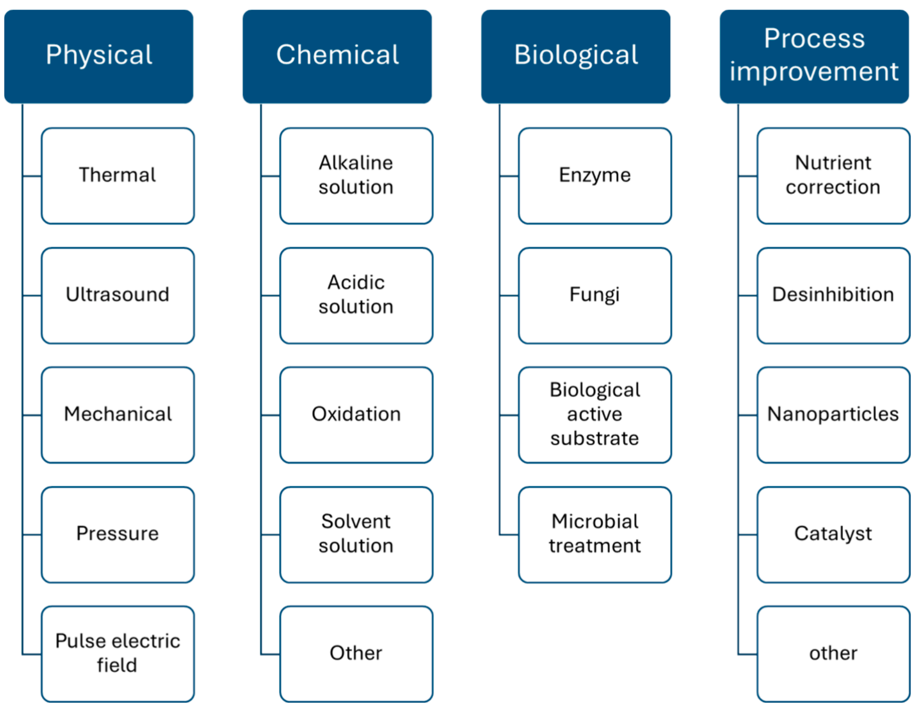

Fig. 3. Diagram of different processes found for pretreatment.

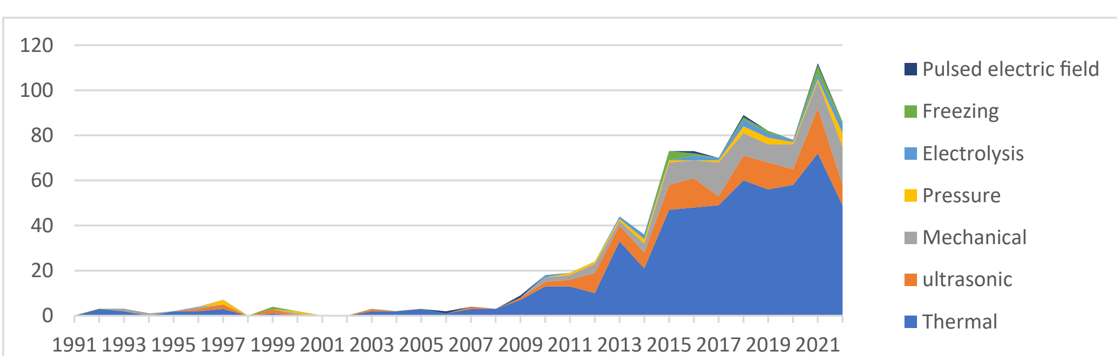

Fig. 4. Count of references of physical methods from 1991 to 2022.

environment like, for example, autoclave.

Pulsed electric field is a new method with the first reference found in 2016. It employs a sudden burst of electric discharge to create a shockwave, radiation and radicals that can disrupt cells[26,118].

3.2. Chemical methods

Chemical methods use different types of chemical compounds to increase the bioavailability of the substrate. As shown in Fig. 5, it is dominated by 2 main groups, alkaline (396) and acidic(138) compounds. In recent years, oxidisers (96) and solvents (45) have attracted increasing interest.

Alkaline and acidic compounds aim at modifying the pH to break down material. Alkaline compounds degrade mainly lignin and require only mild conditions with non-polluting compounds [58]. The compounds found were mainly NaOH but also ammonia, CaO, CaOH, KoH, $CaCO_3$, and black liquor, an alkaline waste from the paper industry. Acidic compounds can be corrosive and toxic, but they are very effective in hydrolysing cellulose. The acids observed were H_2SO_4, HCl, citric acid, free nitrous acid, H_3PO_4, organic acids (lactic, acetic, butyric and oxalic acids), as well as waste gas extracted from waste product combustion and vinegar residue. CO_2 is also used in certain cases to decrease pH through pressurisation.

Oxidisers can be used to break down bonds to break down a structure such as lignin [117]. Oxidisers are mainly referenced under the form of peroxide (H_2O_2, CaO, and peracetic acid), as well as ozone·H_2O_2 is often used under alkaline conditions and has been shown to be effective in removing lignin and hemicellulose [8]. Some other techniques involve wet oxidation, K_2FeO_4, fenton, hypochlorite, chlorination, potassium ferrate, peroxymonosulfate and, polyoxometalates. Some other techniques can be qualified as advanced oxidation process, as they generate OH radicals that can further degrade organic material due to its high reactivity [98]. This process includes peroxide in tandem with UV and/or Fenton (H_2O_2 + Fe). Photooxidation, using mainly (TiO_2), is also a method employed. Gamma radiation can also be used as an indirect oxidiser that generates oxidising radicals [39]. Strictly speaking, this method should be included in physical methods as there is no input of any compound, but since this is a minor method and mentioned as an advanced oxidation process, it is included in this category.

Solvents are used to dissolve recalcitrant matter such as lignin and cellulose to increase bioavailability [40]. Components observed were organosolv methods, ionic liquids, *N*-Methylmorpholine *N*-oxide (NMMO), deep eutectic solvent, ethanol, acetone.

Other techniques observed included the use of surfactants, deodorants, solubilizers, and salts.

3.3. Biological methods

Biological methods use the activity of living organisms to perform the pretreatment using different methods (Fig. 6). Microbial treatment can come in different ways, using specific bacteria or microbial consortia, in an aerobic or anaerobic environment. Other techniques involve the use of fungi directly or through extracted enzymes. The last category observed involves the use of biologically active substrates to break down the substrate, this can be through the reuse of digestate, adding compost, rumen, or activated sludge.

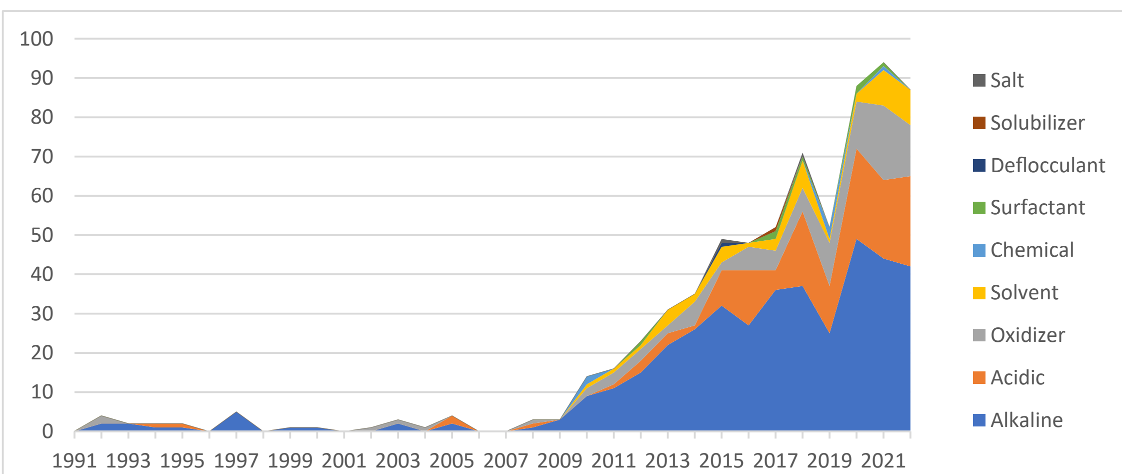

Fig. 5. Count of chemicals methods found from 1991 to 2022.

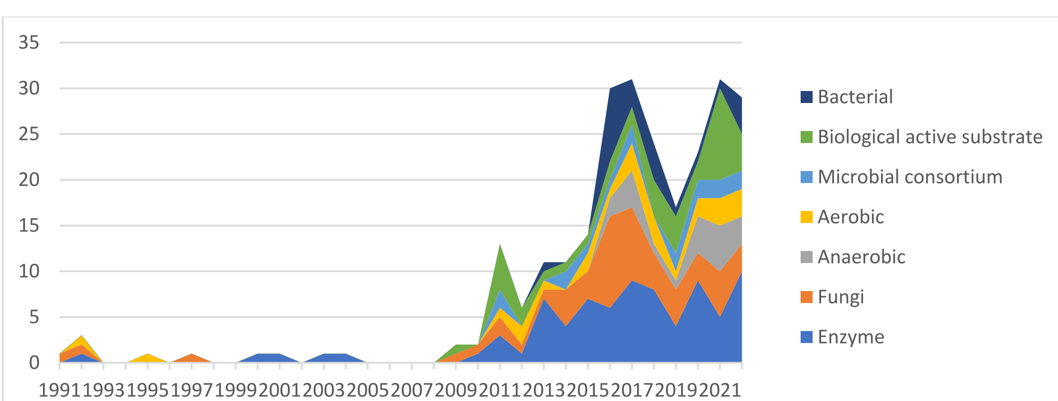

Fig. 6. Count of Biological methods from 1991 to 2022.

3.4. Process improvement

The last category with the lowest number of studies is those methods that do not fit into any other category. They do not directly aim to increase bioavailability through the solubilisation or breakdown of the material, but instead have an indirect effect or help stabilise the process (Fig. 7). There is, for example, the use of nanoparticles that increase the methane production [134]. These nanoparticles may be important as a micronutrient supplement in certain cases [101]and to improve the destruction of the cell wall by microorganisms [135]. Nutrient supplements are also mentioned with supplements such as: urea, ammonia, calcium, iron, nitrogen, sulphur, polyhydroxyalkanoates, sulphate, and trace metals. For disinhibition or reduction of the effect of the toxic compound, different methods were found. Activated carbon or biochar can reduce the effect of toxic compounds [38]. Ammonia removal can reduce its inhibition effect[141]. The coagulant serves to remove toxic metals [2]. Lastly, lipid extraction is applied to algae to use lipids for other purposes but can be considered a pretreatment due to the positive effect it has on biogas production [85]. Filtration can also be used to remove toxic pollutants and suspended solids from the substrate [22].

3.5. Reviews

For the reviews, 38 articles were found that talked about a pretreatment method, while most of the articles focused more on specific substrates. Physical methods were the most predominant with 24 reviews, mostly general reviews, but some more specific about mechanical methods, temperatures, wet explosion, ultrasound, and high-pressure homogenisation. In contrary to the direct analysis of the literature, more reviews were found for biological methods (10) than for chemical methods (4). No reviews covering other pretreatment techniques were found.

The substrate covered by the reviews was mainly lignocellulose substrate with 54 references found (Table 1), some articles were more specific covering a specific substrate type such as wood, manure, or crop waste. Algae was the second most found substrate with 15 references. Other reviews covering a wide panel of different types of waste were found, with a special mention of the potential of plastic (Table 1).

Table 1
Substrates covered by pretreatment reviews.

Substrate	Number of references
Lignocellulose	54
Algae	15
Food waste	9
Sewage	8
Wastewaters	7
Municipal solid waste	5
Palm oil mill effluent	3
High solids waste	2
Waste activated sludge	2
Plastic	1
Oil	1
Paper sludge	1
Mushroom waste	1

4. Reactor design

reactors must increase contact between the microorganism and the substrate to improve digestion and reduce the effect of inhibitors [96]. To this end, there are a large variety of reactors. Some reactors are designed specifically for dry digestion with a reduced water content to reduce size requirement [65]. Other reactors are designed to tackle wastewater and usually employ high-rate reactors [120]. The high rate indicates that the solid retention time (SRT) is decoupled from the hydraulic retention time. The term high rate is sometimes used for other high-performance reactors, but for this review and to keep an objective, rigorous definition, the former will be used.

From 1990 to 2022, 923 articles were found. 204 were rejected, with 1 duplicate. Different categories for rejection were observed: reactors used for other purposes: hydrogen, ethanol, or algae production, gas upgrading reactors, H2S or ammonia removal and pretreatment reactors. Unfortunately, some articles are also rejected due to a lack of information and no access to the article. 43 review articles were found.

676 articles remained and were reviewed. A large variety of reactors design were found and divided into categories according to their operating procedure. 742 different reactor references were found, with all high-rate reactors combined into one group, making the largest group researched (Fig. 8).

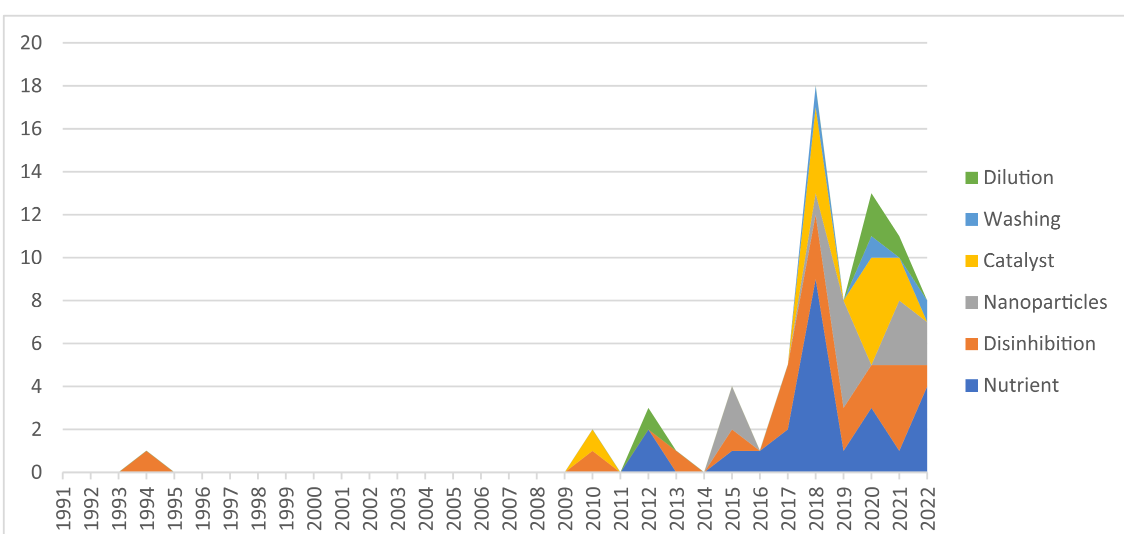

Fig. 7. Count of process improvement methods from 1991 to 2022.

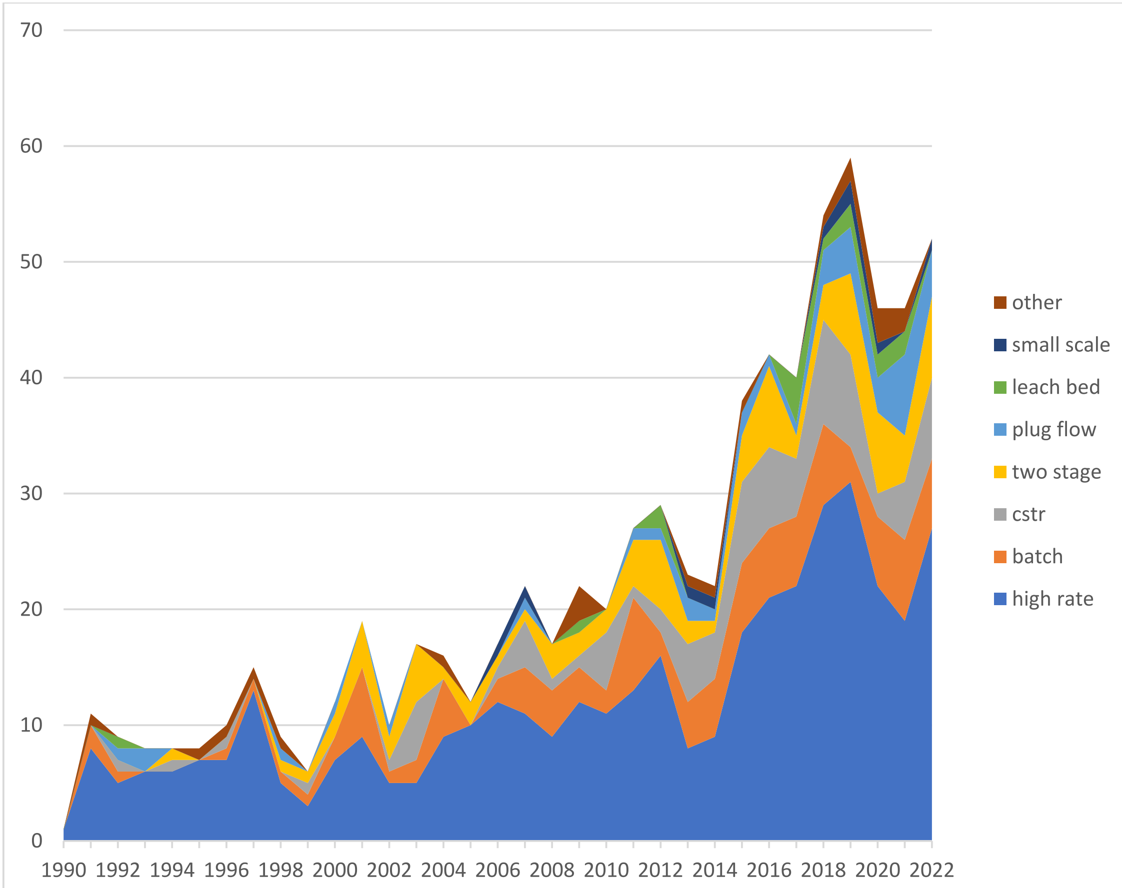

Fig. 8. Evolution of reactor references from 1990 to 2022.

4.1. Batch

A batch reactor is probably the simplest one, with a container filled by the substrate and removed when the digestion is complete. This reactor is very popular mainly for laboratory experiments, due to its simplicity. Some other reactor designs might employ the batch procedure. 101 references to this digester were found.

4.1.1. SBR

The sequential batch reactor, or SBR, has most of the occurrences with 59 references to it. It works by operating the reactor in a sequence of processes[128]. Typically: filling, reaction, settling and withdrawal with an optional resting period (Fig. 9).

The actual layout of an SBR can vary a good approximation can be resumed to 3 main components: an entrance for the influents, an exit for the biogas and an exit for the effluents (see Fig. 9).

4.1.2. Leach bed

Leach bed reactor is a type of batch reactor for solid substrate. It is sprinkled and the percolation or leachate is collected through a mesh, that is then used for recirculation (Fig. 10). 15 references to this design were found.

4.2. CSTR

The continuous stirred tank reactor, or CSTR, is the simplest design of a continuous reactor with 83 observations in the literature. They are mixed reactors that are continuously fed and emptied [24]. They usually consist of an entrance for the substrate, an exit for the gas, an exit for the digestate, and a mixing device (Fig. 11).

For CSTR, one experiment with a rotating reactor was observed in which the whole reactor rotates to increase the mixing in dry digestion [94].

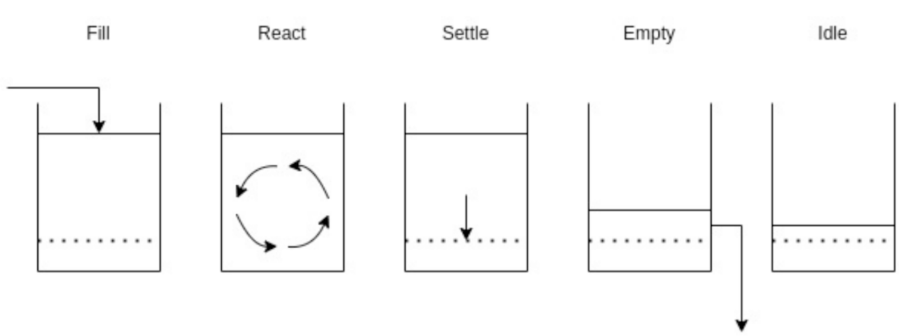

Fig. 9. Operation phases of a SBR cycle inspired by [128].

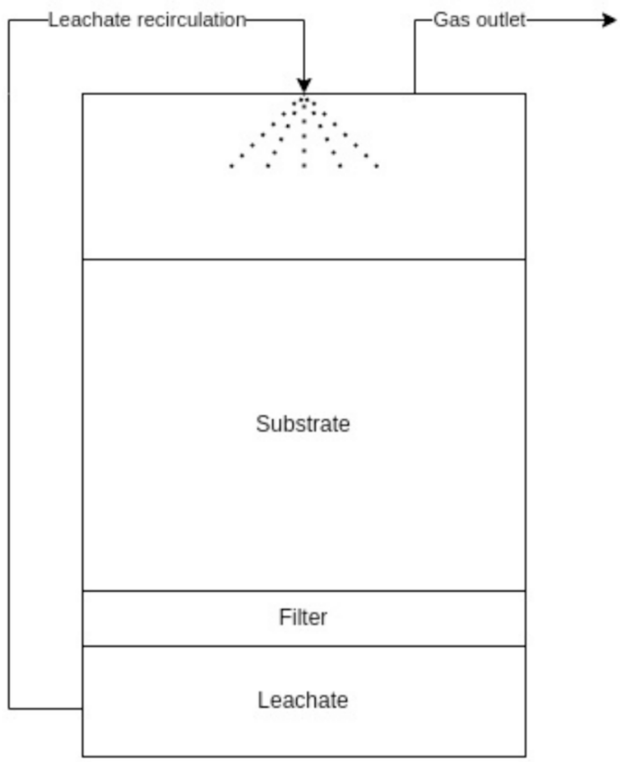

Fig. 10. Leach bed reactor schematic design, inspired by [103].

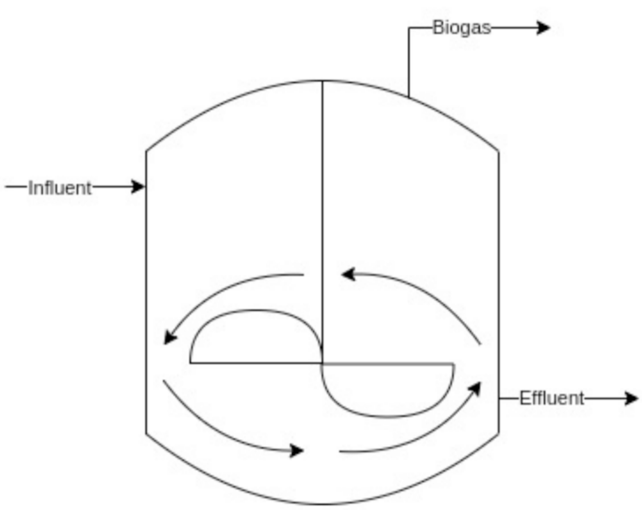

Fig. 11. CSTR schematic diagram, inspired by [95].

4.3. Plug-flow

Plug flow are also simple reactors, less popular in the literature than CSTR with 37 references. In theory, a plug flow is defined as a reactor in which the material moves from one end of the reactor to the other without mixing with the material before or behind it. This ensure that all substrate stays the same amount of time in the reactor. It is often used for dry digestion [33]. As shown in (Fig. 12) there is often a recirculation from the digestate to the fresh substrate for inoculation.

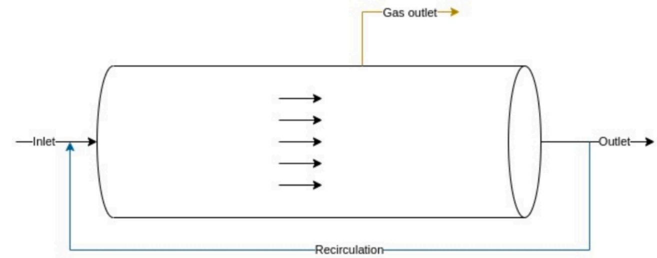

Fig. 12. Plug flow schematic diagram.

4.4. High rate

As mentioned above, high rate is defined as the decoupling of SRT from HRT, and this process is used for wastewater type substrates with a high-water content. This helps to reduce the size requirement. It is the most popular in the research field, with 396 references of 742 total references found.

4.4.1. Sludge retention reactors

Sludge retention can be defined as the settlement of sludge inside the reactor by sedimentation or the formation of granules.

4.4.1.1. Anaerobic contact reactors. Contact reactors are a combination of a CSTR with a sedimentation tank (Fig. 13). The settled sludge is recirculated back while the liquid effluent is removed[82]. This is the simplest design older than the UASB and has seen wide usage in industrial case [111], but only 3 mentions were found in scientific work.

4.4.1.2. Upflow anaerobic sludge bed (UASB). The UASB is a mature wastewater treatment technology with good cost-effectiveness and a reduced footprint compared to other technologies, introduced in the 1980 s [25]. This can be confirmed with the number of references observed (146). UASB uses the natural formation of granules from microorganisms to form a sludge bed together with a gas–liquid-solid separator at the top [125]. The influent is pushed through the granulated sludge and then pushed upward, where the unsettled biomass can coagulate to settle back down. The flow through the sludge helps to prevent any clogging [25], while also allowing for good contact between substrate and biomass. Subsequently, the separator contains a baffle-like structure to redirect the gas to the cone collector and separate it from the effluent [99]. The different layouts can be seen in Fig. 14.

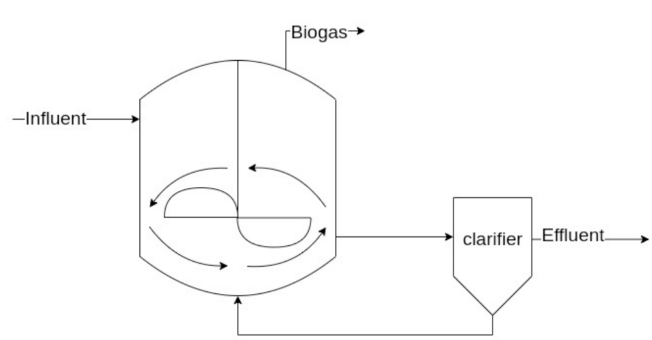

Fig. 13. Schematic diagram of the contact reactor, inspired by [60].

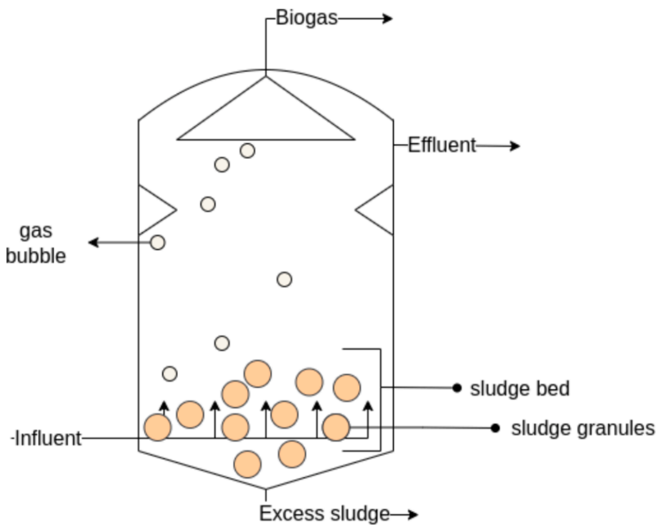

Fig. 14. Schematic design of a UASB reactor, inspired by [99].

4.4.1.3. Expanded granular sludge bed (EGSB). It was developed to solve the issue of not ideal mixing in the UASB, which produced dead zones. The implementation of recirculation and taller reactors led to the development of EGSB. It has a higher velocity, which fluidizes or expands the sludge bed, improving mixing and increasing contact with the wastewater. This increases the efficiency of the digester and makes it more desirable for pollutants, but the higher velocity will wash out suspended solids [111]. Therefore, the design of the EGSB is very similar to that of the UASB, the difference being taller dimensions and the use of recirculation (Fig. 15). However, it is not as prominent in research with only 20 references found.

4.4.1.4. Other UASB derivatives

4.4.1.4.1. Internal circulation reactor. The internal circulation reactor is another evolution of the UASB, it can be described simply as two UASB stacked on top of each other (Fig. 16). Its innovation characteristics is reflected in the literature with 5 references found and the

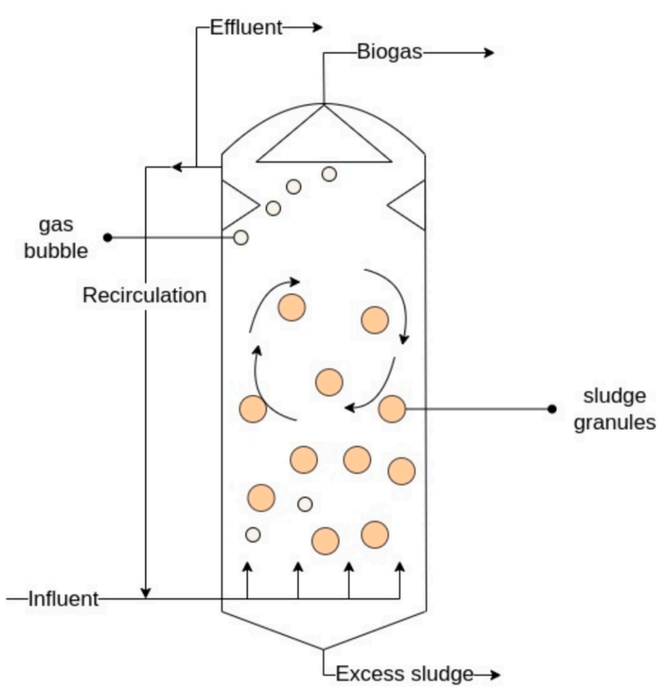

Fig. 15. Schematic design of an EGSB based on [111].

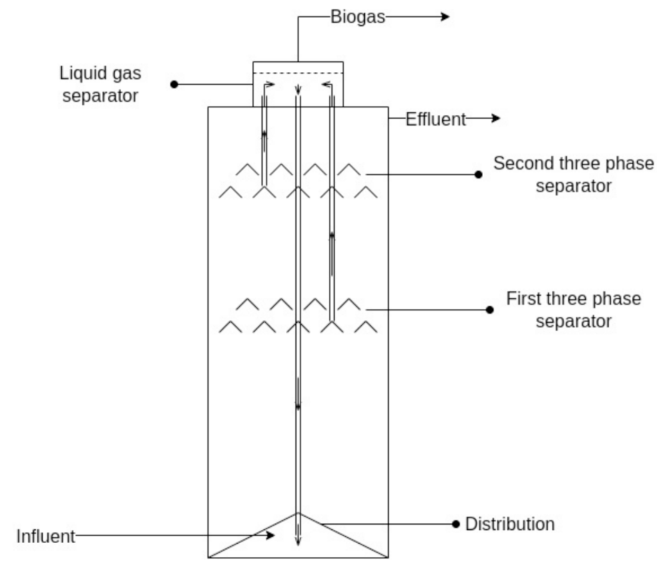

Fig. 16. Schematic diagram of an internal circulation reactor, inspired by [48].

first one in 2015. It is mentioned as a third-generation digester that succeeds the second-generation UASB reactor [72]. The name comes from the recirculation of biogas through the digester[55]. The lift effect also leads to some researchers calling this reactor under the biogas lift reactor name[131]. However, this name is also used for other biodigesters that use biogas recirculation [34], as well as for a small-scale digester design [15]. An internal reactor works with a high rate first stage where the produced biogas is lifted with some liquid, this is then separated at the top where the biogas is recovered, and the liquid is pushed back down to achieve greater flow rate. The second stage is present to treat wastewater with lower organic load (G. Y. [137] Fig. 9.

4.4.1.4.2. Compartmentalized anaerobic digester. A patented digester currently in laboratory size, but according to its creators, has the potential to support a much larger OLR than other UASB and derivates [52]. Details are lacking, but it works by dividing the reactor into 3 parts. It has a distribution zone, a compartmentalised reaction chamber in the middle and a separator at the top.

4.4.1.4.3. Induced bed reactor. Another type of UASB derivative was developed for substrates with a high solid content. The design is patented [28]. It works similarly to an UASB, the main differential parameter is the auger and the septum baffle, as well as the diffuser plate (Fig. 17). It has received some attention with 5 references found but only from 2010 to 2014.

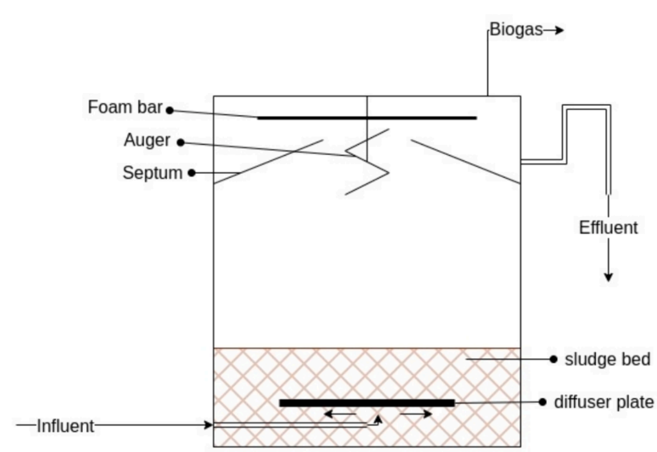

Fig. 17. Schematic diagram of an induced bed reactor, inspired by [35].

4.4.1.4.4. Upflow solid reactor. It is a digester that is common in China, as a simplified UASB reactor with good properties for a higher solid content [64] see Fig. 18. It has seen little research with only 3 references, albeit quite recently(2017,2019,2020).

4.4.1.5. Baffled reactor. A baffled reactor is a type of simple reactor with sludge retention; unlike UASB and derivatives, there is no use of separators but a series of vertical baffles that force the flow up and down through the different baffles (Fig. 19). Its relative simplicity have made it quite well researched with 38 references. As a result of the slow horizontal mobility of the bacteria, this ensures good sludge retention and good contact between substrate and microorganisms. Its functioning can be modeled as a fixed film [9]. It also has the advantage of separating acidogenesis and methanogenesis into different compartments [12].

Many configurations are possible[12]. Next to the traditional design the following configurations were also observed in this study:

The periodic baffled reactor (2). It works with a circular reactor and incorporates the substrate in the different baffles sequentially to ensure homogeneous load across the digester and has the presence of methanogens even in the first baffle [115].

One mention was found of a vertical baffle reactor[29].

Lastly, the inclined baffled reactor (3) is a modification of the baffled reactor to increase solid retention by adding a more complex flow route to improve mixing and contact.

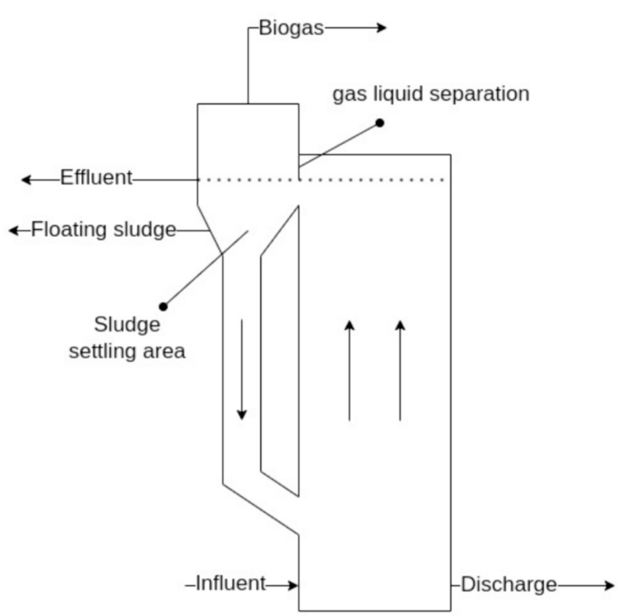

Fig. 18. Schematic diagram of an upflow solid reactor, inspired by [64].

4.4.2. Biofilm reactors

This is a broad category, refers to a reactor with the use of a media where biomass can be retained and offers space for a biofilm to grow [4,27]. They can use a wide variety of packing materials ideally porous, as they can be prone to accumulation of nonbiodegradable material they are more suitable for diluted wastewater[100]. However, they have an advantage in simplicity[27].

4.4.2.1. Anaerobic filter reactors (10) and fixed bed reactors (31). Fixed bed or anaerobic filters refer to biofilm reactors where the media is settled in the digester (Fig. 20, left). The fixed film (11) generally refers to the same concept. Some studies use textile as fixed film such as in the down-flow stationary fixed film reactor [129]. Some other references to the same concept of a reactor filled with a media include packed bed(18) or fixed bed (1).

4.4.2.2. Fluidized bed (37). Specific type of biofilm reactor where the support media is in suspension in the digestate or fluidized (Fig. 20, right) thanks to the upflow drag forces[81]. It operates by a fluid rate carefully considered to ensure fluidization without washout[5].

4.4.2.3. Alternative configurations (inverse, horizontal). Horizontal reactors with plug flow characteristics[75]were also observed with 6 references.

It can also be found in downflow configuration, with 7 references in the observed literature, with a floating media. In the case of the fluidized bed, it has the advantage of low energy needs for fluidisation and no settler needs [42]. These types of digesters are often called inverse bed [19,42].

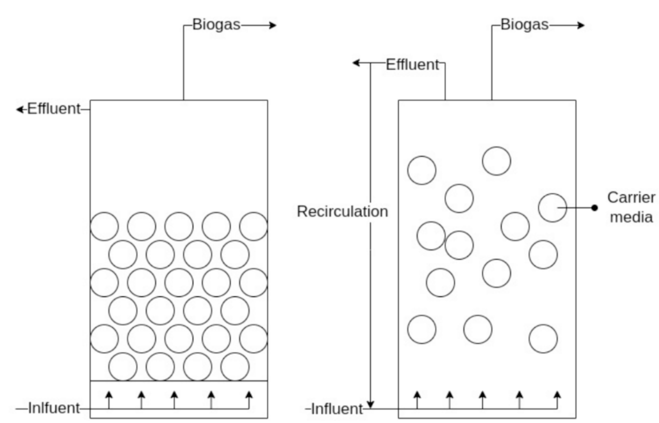

Fig. 20. Schematic diagram of a fixed bed (left) and a fluidized bed (right).

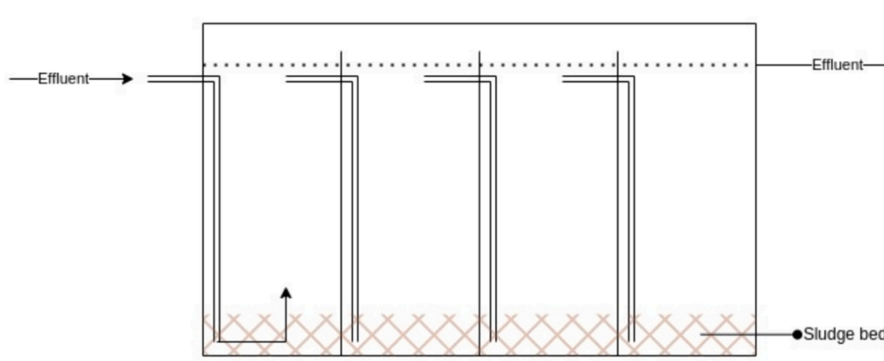

Fig. 19. Schematic diagram of a baffled reactor, inspired by [36].

4.4.2.4. Moving bed (2). Another type of biofilm reactor, but instead of using a unidirectional flow it operates more like a CSTR. It is a mixed reactor filled with a carrier [82].

4.4.3. Membrane reactor (6)

These relatively new types of reactors use a membrane to retain solids in the digesters and have seen sporadic research from 2009 to 2022. This concept can help to ensure complete digestion, by completely separating solid retention time from HRT, due to the filtration effect of the membrane [23]. It can be useful in extreme scenarios where sludge retention does not occur properly due to a lack of granulation [51]. Different configurations can also be used with a membrane inside the reactor or outside the reactor [23].

4.5. Hybrid reactors

Hybrid reactors are reactors that combine the properties of multiple reactors. The most common, often referred to as a hybrid reactor, is a UASB with a fixed bed integrated. 52 references of hybrid reactors starting in 1991, meaning it is not a new idea. Other configurations exist, to combine a high-rate reactor with a fixed bed such as the EGSB and baffle reactor. A mention of sequencing batch reactor with a biofilm carrier was also observed.

A combination of different compartments is also often called a hybrid reactor, with examples of fixed and fluidized bed combinations with different flows (up and down)[105].

The use of hybrid reactors seems to be somewhat overused and abused, with often no clear indication of what they are hybrid of. As seen, a combination of different compartments is often used to call a reactor a hybrid but would be more appropriately called multistage.

4.6. Multistage reactors

Adding different stages is also popular in the literature with 83 references for two stage reactors and 4 for three stage reactors. These reactors are often a combination of multiple stages of CSTR reactors. Another popular category observed is the combination of a leach bed as the first stage for solid digestion and a high rate second stage. However, a large variety of combinations is possible.

4.7. Small-scale reactors

small-scale reactors are low technology reactors with usually no mixing and heating. They are mainly employed in developing countries usually family scale of a few cubic meters. The specific designs observed in this study are fixed dome reactors (5), with floating drums and prefabricated plastic or tubular reactors also appeared once each. This correctly represent the 3 main branches of small-scale designs (Fig. 21). They all possess their advantages and disadvantages. Fixed dome reactors are long lasting but need good construction to be properly sealed. Floating drums require more maintenance but provide biogas at a constant pressure and plastic digesters are cheap and easy to setup but with a limited lifespan.

4.8. More anecdotal reactors (less than 5 occurrence)

4.8.1. Rotating disk/contactor

Encompass designs where, as the name implies, a disk rotates inside the reactor, allowing for suspended and attached growth (Fig. 22). Offering then a large specific area and are not very sensitive to toxic substances [107]and are for example used in textile industry[66]. They can be used in batch mode[107]or in continuous mode[66].

There is also a mention of a fixed bed disk reactor. Works in the same way as seen before with the disk reactor in the batch system. In this case, it is a vertical upflow digester where the influent passes through the horizontal disks and leaves at the top [121]. Unfortunately, it lacks comparison with other digester types. These types of digesters could, as mentioned in the last example, be qualified as a fixed bed, but in most examples, they are not mentioned as such.

The rotating disk reactor is also mentioned once as a batch reactor.

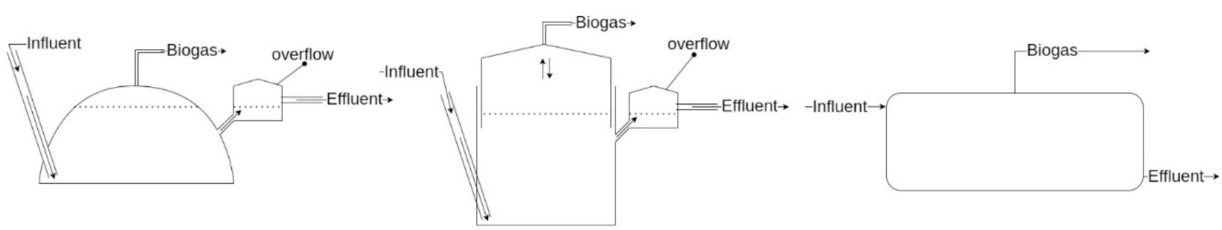

Fig. 21. Schematic design of small scale reactor types, from left to right: fixed dome, floating drum, plastic tube reactor.

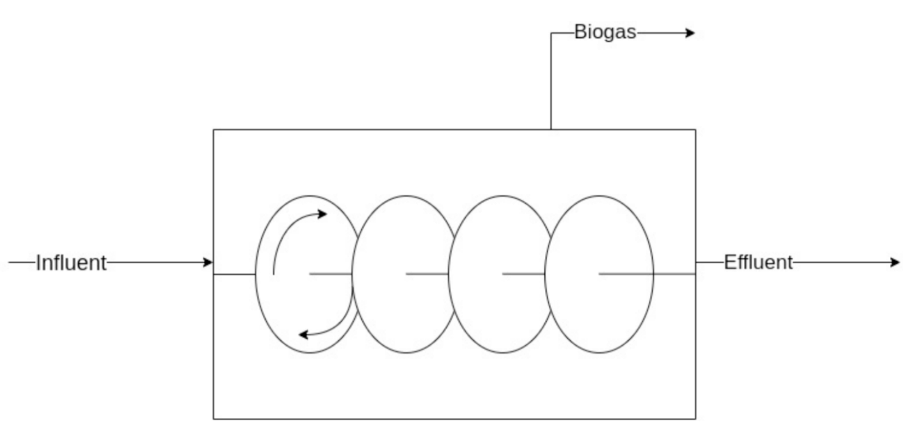

Fig. 22. Rotating disk reactor schematic diagram, inspired by [107].

4.8.2. Gradual concentric chambers reactor

This digester is mentioned once as a cost-effective simple reactor for wastewater treatment for tropical and subtropical areas. It is a reactor than comprise of an anaerobic and an aerobic part (Fig. 23).

4.8.3. Tower

This is an older concept with 2 articles from 1996 and 1998 that only study a model reactor[92,102]. It is a vertical reactor where the gas movement creates a fluid circulation similarly to an airlift reactor, and it is composed of different modules for gas control where excess gas can be removed to ensure homogenous suspension [92,102]. An example can be seen in (Fig. 24).

There is also a mention of a multiplate reactor with the same characteristics, apart from the dimension of the reactor that is wider[44].

4.8.4. Loop reactor

As the name implies, this type of reactor has a physical loop of the substrate through the reactor (Fig. 25); this is traditionally done through gas circulation but can be supplemented by a screw to increase flow and mixing [59]. The author mentions satisfying results for biogas production but is not very popular, only 2 references. It is probably surpassed by high-rate reactors.

4.8.5. Lagoon

Lagoons are simple reactors with long retention time, normally used to treat wastewater or animal waste [130]. They are often not heated, and therefore they are more adapted to warmer climates [110]. The design consists of a large, elongated digester with a simple entrance and exit on either side (Fig. 26). Interestingly it only appeared in 2009, 2019 and 2022 indicating that there is modern interest in simple cost-effective digesters.

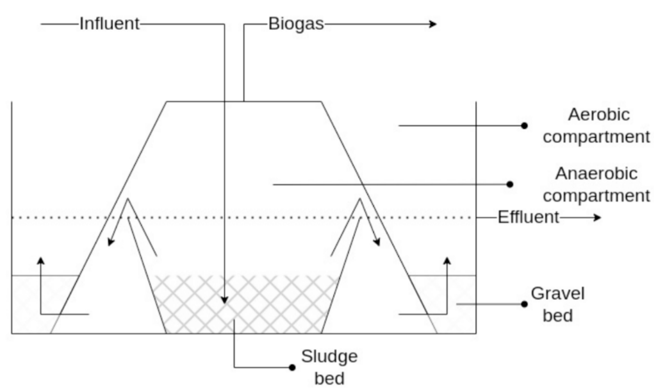

Fig. 23. Schematic diagram from the gradual concentric chambers reactor, inspired by [77].

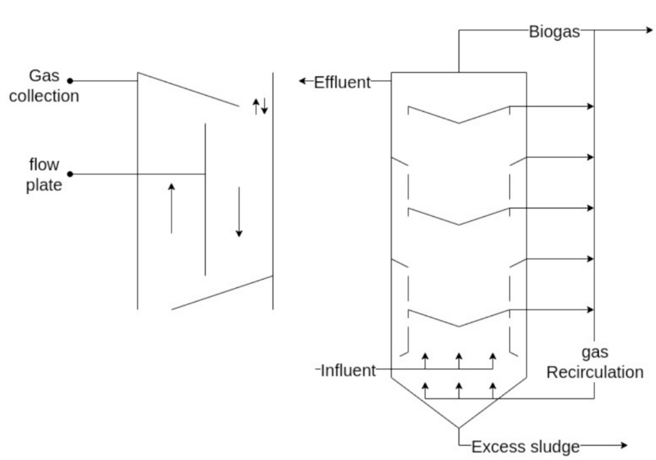

Fig. 24. Tower reactor module (left) and design of a reactor with multiple modules (right), inspired by [74].

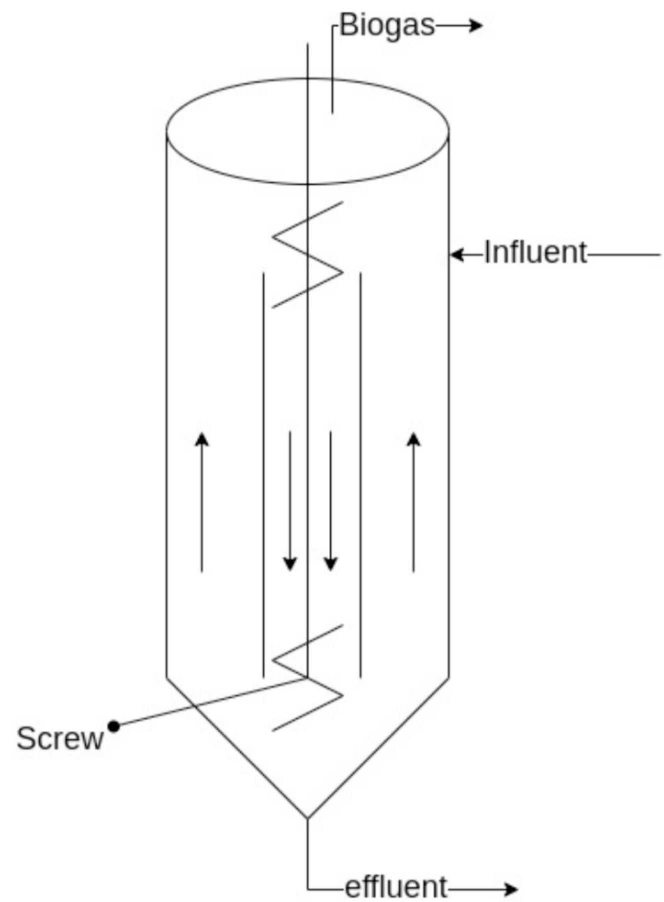

Fig. 25. Loop reactor schematic diagram, inspired by [59].

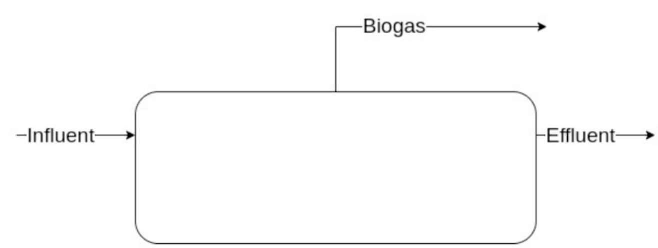

Fig. 26. Lagoon reactor schematic diagram.

4.8.6. Hydraulic flush

A hydraulic flush is a laboratory, mixed reactor operated semi-continuous conditions. It works by removing a part of the digestate and separate the solid and liquid part. The liquid part is sent to a second stage for methane production, while the solid part is returned to the digester with fresh substrate and water. The flush naming comes from the removal of a part of the digestate as waste before refilling the digester with the fresh mix[11]. The main objective behind this digester is to reduce ammonia inhibition form slaughterhouse waste and to perform research on a second stage in a controlled environment [11,127].

4.8.7. Bionic reactor

The bionic reactor is a type of reactor design that aims to imitate animal digestion strategies as they are more efficient and faster at digesting complex substrate. There is no specific design for this but

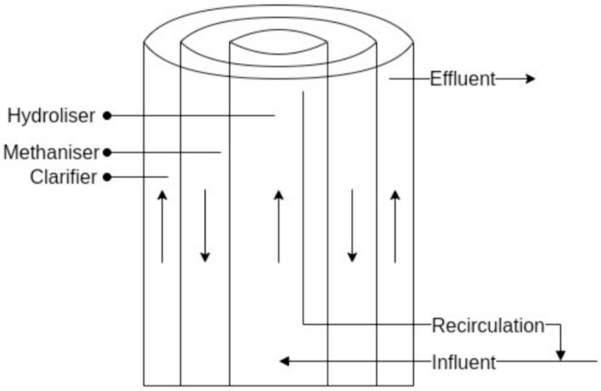

Fig. 27. Schematic diagram of a labyrinth flow reactor, inspired by [31].

rather different path of improvement such as inclusion of multiple stages, bioaugmentation with protozoa and/or fungi [13]. The specific study found tried to imitate the rumen of cattle [138].

4.8.8. Labyrinth flow reactor

It is described as a UASB reactor with separation of the acidogenesis and methanogenesis and a settling tank[31]. In practise, it is very similar to a baffled reactor with different compartments and a liquid up- and downflow (Fig. 27).

4.8.9. Self-flotation reactor

The self-flotation reactor was developed for wastewater with large amount of suspended solids[136]. It works as a specifically designed upflow reactor so that biogas bubbles can capture and remove suspended solids (Fig. 28).

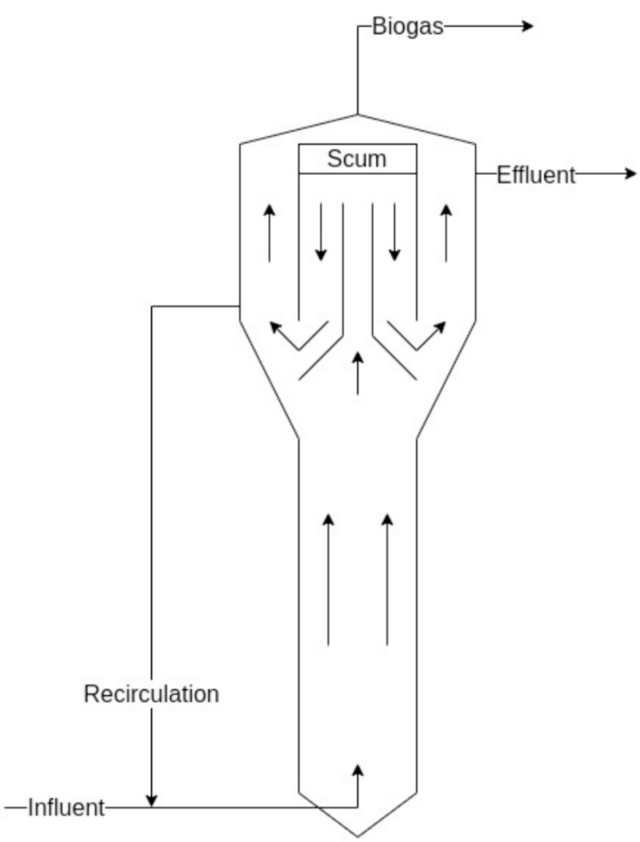

Fig. 28. Schematic diagram of a self-flotation reactor, inspired by [136].

Table 2
Reactors sizes and number of references.

Type	Size	Number of references
Full scale	>10 m3	44
Pilot scale	>100 Liters < 10 m3	80
Laboratory scale	< 100 Liters	492
"Small scale"	/	7
Model	/	50

4.9. Reactor sizing

In terms of sizes, next to the 3 predefined categories, 2 more categories needed to be added, namely small-scale reactors, that refer to the class discussed before of low tech solutions, and models (Table 2). Lab scale reactors compose the vast majority of the references found, being 6 and 11 times more prevalent than pilot and full-scale reactors. A few articles mentioned small scale reactors type. Moreover, there were 50 references to mathematical model reactors such as the ADM1 model.

4.10. Reviews

Of the 43 articles found; 9 had to be excluded as they are not reviews of design but rather a review of substrate or pretreatment techniques employing a specific type of reactor design. For the reviews left 14 of them were generalists not covering any specific type of reactor. 13 of them were covering high-rate reactors consistent in popularity with the literature search. The other reviews found were 1 for CSTR, 2 for SBR and 1 for two-stage reactors. Lastly, 3 articles covered solid-state digestion.

5. Temperature

Microbial organisms that perform anaerobic digestion are temperature dependent, with optimums at 37 °C for mesophilic digestion and 55 °C for thermophilic digestion[113]. More temperature ranges can be used, such as psychrophilic and hyperthermophilic.

From 1991 to 2022, 907 articles were found, 37 were rejected, with 4 duplicates. Other reasons for rejecting were consideration of temperature in pretreatment or biogas upgrading, the study of reactor performance parameters such as acetate or foaming, reactor designed for hydrogen production, and pathogen study, which are also interesting parameters to consider, but outside of this scope. 21 more reviews were not considered in further research. In total 849 articles, with a research focus on temperature were reviewed (Fig. 29).

The thermophilic range has the most research with 587 mentions, 33 of them are in the hyperthermophilic range and 554 in the more usual thermophilic range. Mesophilic range has 446 mentions. Although mesophilic is in practice more common, this can be indicative of the potential of thermophilic digestion and the research needed to perform it in a stable way[106]. The Psychrophilic range is also researched in lesser numbers with a relatively low 88 articles found but it is not a new research area as the first mention is in 1994. The research concept might have to evolve with a proposal to divide psychrophilic in 2 categories with psychotrophic around 20 °C and true psychrophilic around 10 °C [3].

There is a large diversity of combination tried in multistage reactors, 51 cases were observed. The majority of the cases are from thermophilic to mesophilic (31), a smaller group from mesophilic to thermophilic (9). The combination was also present with hyperthermophilic temperatures: from hyperhermophilic to mesophilic (4) and from mesophilic to hyperthermophilic (1). Two cases were found from hyperthermophilic to thermophilic (2) and 1 for the opposite. One last experiment used tree stages with a switch from mesophilic to thermophilic and back.

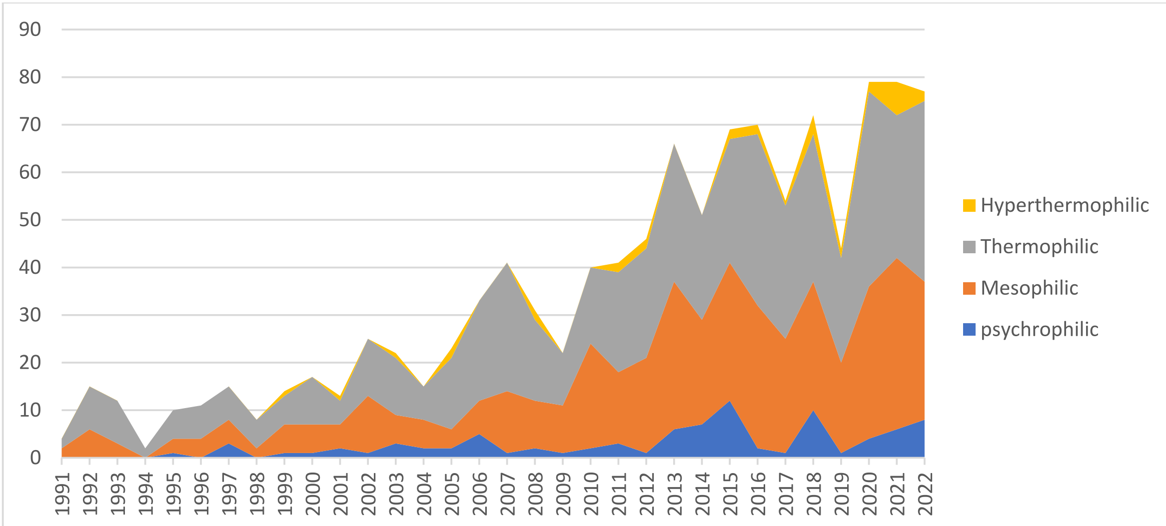

Fig. 29. Evolution of temperature research from 1991 to 2022.

5.1. Reviews

Thermophilic digestion, with 12 references found, was the most reviewed. More reviews were found for the Psychrophilic range (9) than for the mesophilic range (6), this is not reflected in the literature search where psychrophilic is much more anecdotal. The number of reviews covering the psychrophilic range is surprising considering that only 88 references were found. No reviews were found that specifically covered the hyperthermophilic temperature range. However, some reviews would cover multiple ranges.

6. Biogas upgrading

The biogas produced is mainly composed of methane +/-60 % and CO_2 +/-40 %. Biogas can be upgraded to biomethane by reducing the CO_2 content; in the literature, processes can be divided into physical, chemical, and biological [7].

The total number of studies found from 1998 to 2022 was 658. Of those, 94 were rejected for reasons such as: covering other impurities (H2S, siloxanes), of topic, other use of biogas, or a lack of information. 94 more articles were reviews. Of those 470 studies were left for further study (Fig. 30).

The biggest group found was for sorption methods with 183 references, this group was divided in absorption and adsorption.

6.1. Absorption

Absorption was found in 112 references. Absorption is defined as a gas to be dissolve in a solvent, based on solubility to absorb co2 selectively[119], or by a chemical by reaction[61].

Found examples were:

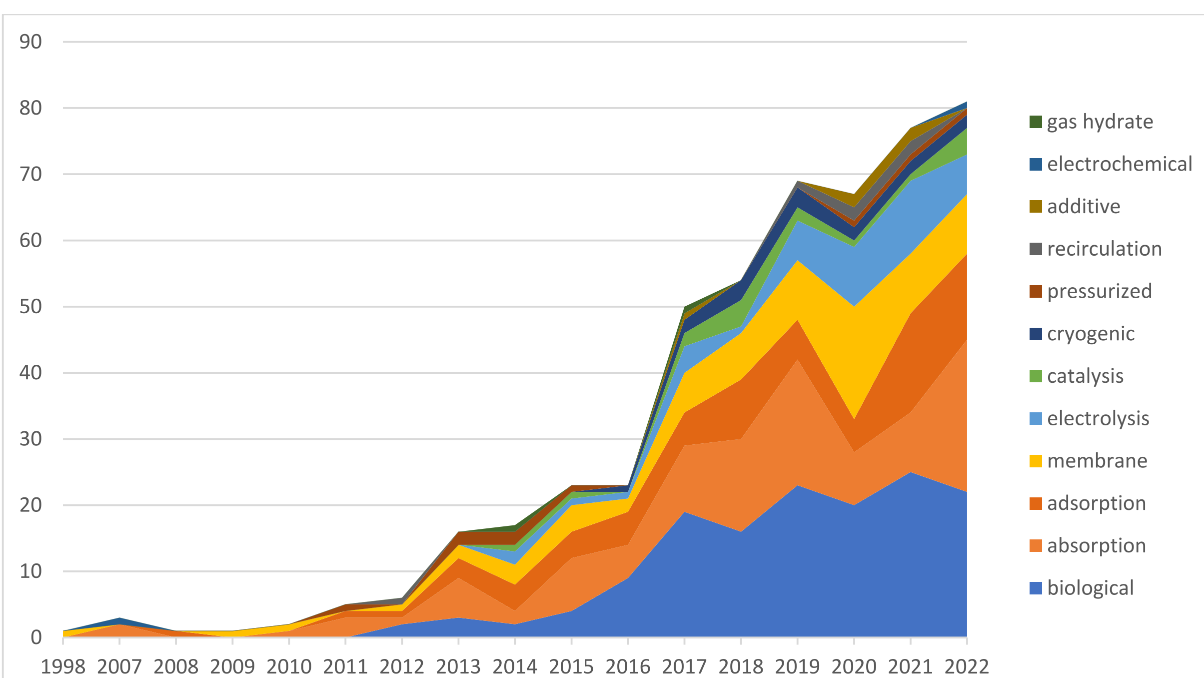

Fig. 30. Evolution of biogas upgrading methods from 1998 to 2022.

- water (46) with one case of seawater,
- amine compounds (40) different types used were: MEA (ethanoloamine), MDEA (Methyldiethanolamine), DEA (diethanoloamine), 3-AP (aminopropanal), AEEA (Aminoethylethanolamine), DEIBA (2,2′-Dibenzimidazole), AMP (2-amino-2-methyl-1-propanol), DGA (Diethylene Glycol Amine), TEA (Triethanolamine), MMEA (Monomethyl Ethanolamine), PZ (propylamine)
- alkaline (32): the specific form is not always mentioned but mainly NaOH. Also, K_2CO_3, KOH, $NaHCO_3$, $CaOH_2$ and NH3. Some waste materials were encountered under the form of ash and air pollution control residue.
- ionic liquid (8), under the form of: BMIM (−Butyl-3-methylimidazolium), BMIM Br (1-butyl-3-methylimidazolium bromide), AMIM formate, imidazolium, PMIM br(1-propylamine-3-methylimidazolium bromide), OMIM BF4 (1-octyl-3-methylimidazolium tetrafluoroborate)
- deep eutectic solvent (DES) (8): mentioned as Choline chloride or sometimes as choline chloride/Urea.
- Other substrate found with less references were: fe/EDTA, amino acids, CaCl2, wastewater, PEG and, porous liquid.

6.2. Adsorption

Adsorption was found in 72 references: It is defined by a physical bind on the surface of solid. In research there is a focus on surface area and selectivity as well as regeneration ability[63].

Found examples were:

- carbon (25) from multiple sources: biochar, hydrochar, activated carbon (specific waste source for it are: seeds, wood pellets, coconut, leather industry), carbon molecular sieve with 1 example derived from bamboo.
- Mineral (17) mainly from zeolite, but also olivine, silicate, lewatita, clay, hydrotalcite. Zeolite from natural origin such as tuff or synthetic (13X,4A,5A, bla421-A,Na10K21-A, NaKA,SAPO-56)
- Ash (9) form wood, municipal solid waste or a case of coal fly ash.
- Silica gel (6)
- Waste material (5) from varied sources: construction tuff, industrial steel slag, spent coffee.
- Metal organic framework (4) with the following names: UIO-66, aluminium metal ions, tetracarboxylate ligands, amino-MIL-53, IL@ZIF-8.
- Resin material (3): polymeric, PEI/HP20, polyethyleneimine impregnated NKA-9.
- Other components observed with less references were: clathrate, metal carbides, porous polymeric beads and, xerogel.

6.3. Biological

Biological (1 4 5) methods were divided in 2 main categories: first, the use of hydrogen (82) to remove Co2 while producing methane. This process can be in situ[71,70]or in a dedicated biological reactor[70,71]. Secondly, the use of a photobioreactor (67) to eliminate CO_2, they sometimes mention the use of a high-rate algal pond. Fermentation is another less employed category (3) but it is a recent approach (2018,2019,2023). It is similar to hydrogen methods; it is expected that instead of producing methane, it aims at the fermentation of different products: acetate [89,90], or in the most recent study also ethanol[47]. A reference was found performing a different test with hydrogenotrophic methanogens in situ to improve methane production[93], not a complete removal is observed with only 11 % reduction but notable for just improving the AD process without further needs.

6.4. Membrane

Membrane separation (72) is also a popular method. Large variety of methods, often mention the use of hollow fibre membranes (16) a specific type of geometry popular for biogas upgrading [69].

6.5. Electrolysis

Electrolysis (41) is also a popular method. The concept revolves around the use of an electric current and CO_2 to produce methane, aided by biological activity [16]. Nearly half of the studies (15) were conducted in situ. It can be found under different names bioelectrochemical system, microbial electrolysis cell, electromethanogenesis, and microbial electrosynthesis.

6.6. Catalysis

Catalysis (16) also uses hydrogen but instead of biological transformation uses metal catalysers to do the conversion into methane, through the Sabatier process[53]. Different metal catalysers were tested.

6.7. Cryogenic

Cryogenic (14), uses the different properties of condensation to separate methane from CO_2, where high purity methane and liquified CO_2 are produced [132].

6.8. Pressurized

Pressurized (9) techniques, often called Autogenerative High Pressure Digestion, use gas generated to elevate pressure inside the digester. This create a separation mechanism through higher co2 dissolution potential compared to methane[62]. At 90 bar the methane purity can go as high as 90 %[67].

6.9. Recirculation

Recirculation (6) are methods in which the biogas is circulated back into the digester, although with a low but significant 10 % increase in the concentration of methane [140]. All those studies are recent in between 2019 and 2021 with 1 other case, a bit older from 2012, where part of the sludge is aerated and recirculated in the digester[87].

6.10. Other

In certain cases, additives were added to the digester to improve its performance and increase the methane concentration. They do not necessarily classify as a biogas upgrading method but as they are mentioned this way and improve methane production and concentration, they were included. The components included were zero valent iron as nanoparticles, biochar, CaCl2 and ammonia nitrogen. Some cases of adsorption could be classified as additives through the in-situ use of olivine or biochar. Another case used silicate to adsorb Co2 to help in pressurized reactor to eliminate surplus of CO_2.

2 studies observed used electrochemical methods, they are a bit different from normal electrolysis as they do not use biological activity to transform Co2 to methane, rather use abiotic cathode to capture and separate CO_2 [80].

Gas hydrate (2) is also a technique observed sparkly, it uses the difference in conditions for hydrate formation to selectively separate CO_2 from methane [21,114].

6.11. Reviews

63 of the reviews found were generalist not covering any specific type of upgrading method. Afterwards, membrane technology showed the largest number of reviews with 12 articles, different from the direct article search where absorption, adsorption and biological method showing a larger amount of research. Interestingly 3 reviews claim to be systematic, albeit only 1 of them explain its methodology.

7. Desulphurization or H_2S removal

The biogas also contains impurities, such as hydrogen sulphur, ammonia, siloxanes, and can contain traces of nitrogen and oxygen. Due to the toxicity and corrosiveness of hydrogen sulphur, it requires special attention, and its concentration should be controlled in almost all uses [7].

The total amount of studies found from 1991 to 2022 was 893. Of those, 357 were rejected, the higher number is due to less specific research parameters. The rejections are for different reasons. There is the reduction of the effect of H_2S on engines and fuel cells. The characteristics of substrate and their related H_2S production as well as codigestion optimisation to reduce its production. Research was also found on other components of biogas: Co2 and siloxanes. Research aimed at producing H_2 was also observed. Lastly, some articles had to be rejected due to a lack of information. Further, 59 articles found were reviews, with 422 articles left for further research (Fig. 31).

Main method are sorption methods. Divided in adsorption and absorption.

7.1. Adsorption

For adsorption (146), the concept is the exact same as for biogas upgrading through the binding to a solid surface.

The following techniques were found:

- Carbon (70), generally activate carbon, but also mention of biochar, carbon nanotubes and graphite.
- Metallic (40), generally iron compounds such as iron oxides, iron sponge or steel wool. In addition, other metals were explored, CU, Mg, M0, Zn, and Al oxides.
- Mineral (21), mainly zeolite but also goethite and magnetite.
- Ash (11) from different sources: wood, municipal solid waste, sewage sludge.
- Waste material (7): eggshell, concrete, oxidized steel chips, steel slag, used tire rubber.
- Sedimentary rock (7): bog iron ore, clay, red rock, diatomite, red soil, sand silica, laterite.
- Metal organic framework (4), mil-101, ZIF-8
- Other adsorbers observed in lesser frequency: amine (grafted onto support such as silicates), caco3, chelate, nanoparticles, resin, sewage sludge.

7.2. Absorption

Absorption (55) is based on the dissolving of a gas in a liquid, the concept is the same as specified before for biogas upgrading. However, in this case, the absorber is selected for its selectivity towards H_2S.

The follow techniques were found:

- Water (10)
- Alkaline (20), most often NaOH is found but there is a large variety of compounds observed: $CaOH_2$, KOH, H_2O_2, Manure, $NaHCO_3$, Na_2CO_3.
- FE/EDTA (8), an iron chelate that oxidizes H2S [73].
- Metallic (4), $FeCl_2$, iron sulphate, iron chelate.
- Acidic (3), H_2SO_4, CH_3COOH.
- Amine (4)
- deep eutectic solvent (2)
- Other compound observed in lesser number: aminoacids, ionic liquid, nanoparticles, recirculation of digestate, sedimentary rock (dolomite, lime).

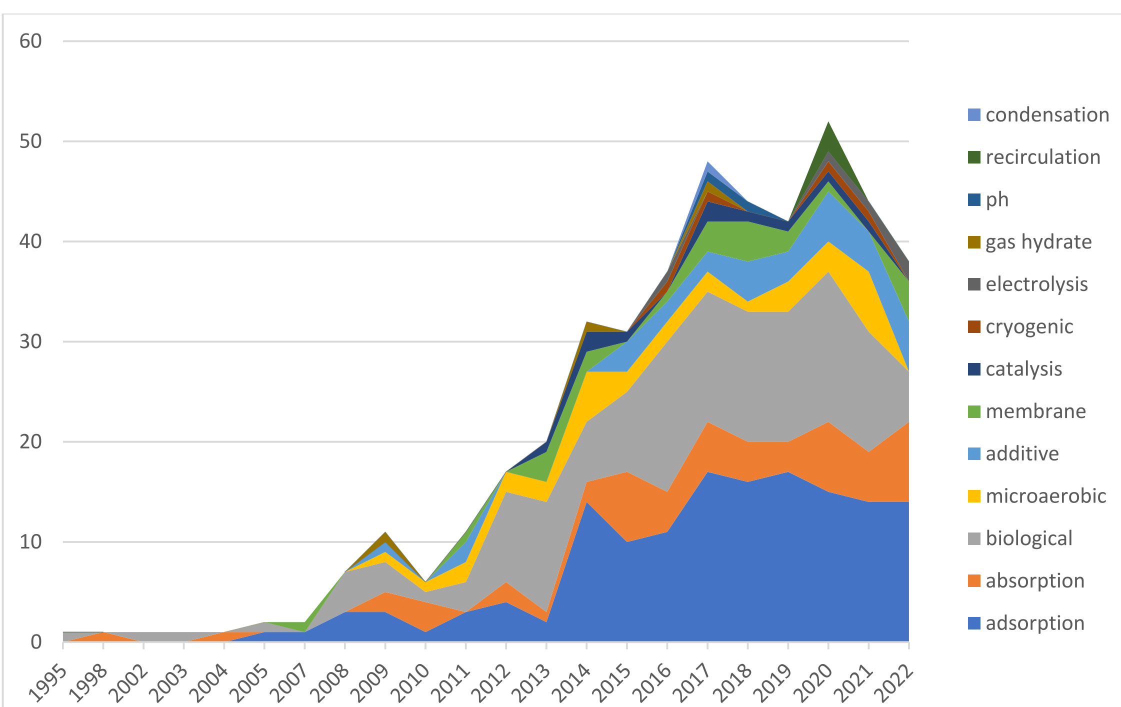

Fig. 31. Evolution of desulphurization methods from 1995 to 2022.

7.3. Biological

Biological methods (1 3 5) were also very present. Different types of bioreactors, mainly biotrickling but also bioscrubber and biofilter were found. Bioscrubber consist of 2 reactors, the first absorb the gas while in the second one the sulfur is oxidized by bacteria. Biotrickling uses packing material and a solution that trickles through the bed, while a biofilter is composed of a carrier to immobilise bacteria with the moist gas that circulates through it [116]. Some mentions of bubble columns were found where the gas flows through a liquid medium containing oxidising bacteria [56]. All of those methods work with the use of sulphur oxidising bacteria. They use air, oxygen, microaerobic or anoxic conditions (nitrate) as an oxidiser.

There were also a few mentions of a photobioreactor and one reactor using purple sulphur bacteria.

7.4. Microaerobic

Microaerobic (32) can technically be considered a biological method, but is considered differently due to the difference in the procedure. Microaerobic works by adding air or oxygen to the reactor to suppress the production of H_2S and oxidize it using bacteria naturally present in the reactor[32].

7.5. Additives

Additives (33) are more prevalent than in biogas upgrading. Large varieties of additives for anaerobic digestion to reduce H_2S generation. Mainly iron compounds were used for this purpose. The different forms found were the following: Fe_2O_3, $FeCl_3$, $FeCl_2$, FeOOH, Fe_2SO, K_2FeO_4 and, zero valent iron. Waste iron was also present from different sources: iron ochre, iron oxide and, iron powder. Mentions of other compounds were found: Carbon (biochar, charcoal), ash, fenton and, NH4Cl.

7.6. Membrane

Membrane methods were also prevalent for desulfurization with 22 references. They also mention the use of hollow fibre structures. Different type of compound were used: polyimide, carbon, Copper sulfate, metal organic framework, polydimethylsiloxane, polyvinylidene-fluoride, polytetrafluoroethylene, zeolite and, ZnO. Also a few mentions of water swollen type and glassy were found.

7.7. Catalysis

Catalysis (11) uses catalytic oxidation using different compounds to oxidize H2S (CeO2/ZrO2 + Ni/Rh-Ni, CeO2-Vanadium, rare earth, Vanadium). Photocatalysis was also found using different compounds: TiO, WO3, NiO in combination with light to oxidize to oxidize H2S[20].

7.8. Other methods

3 examples were found using electricity to oxidise H_2S. two studies uses electrochemical oxidation, this works by absorbing the H_2S and oxidising it and using the electrical current to regenerate the oxidiser [86]. The other study used dielectric discharge to oxidise H_2S by breaking H-S bonds[112].

another technique mentioned is cryogenic (4), uses same procedure as for biogas upgrading[132], but some articles also mention the removal of H_2S with this method[76].

Gas hydrate (3) is also mentioned sparsely for H_2S removal. Some consider biogas upgrading using van der Waals forces to capture molecules in clathrates [114]. The articles mention the capture of H_2S alongside CO_2.

Recirculation (3) is also mentioned in this regard, once with the digestate[83]and twice with the biogas. One study reported a large change in H_2S production[139].

Finally, 2 articles used a process that is a bit different as they use a two-stage reactor to remove the H_2S, the first stage is kept at a pH of 5.5 to produce H_2S to avoid its production in the second stage[123,133]. This process might pose issues as two stage reactors produce H2 and CO_2 in the first stage that can be reused and would be strongly contaminated in this system [70,71].

7.9. Reviews

For desulphurisation, most of the references found were generalist not covering a specific type of methods. Adsorption showed the most interest with 21 articles found. Biological methods showed great interest in the articles research but only 5 references were found in the reviews. Absorption also showed low number of references with 3 reviews articles found. 5 references were found for microaerobic methods, and 2 articles were found for membranes and additives. 2 articles found were systematic reviews covering the technology in general, albeit one is a closed source conference paper.

8. Discussion

In general, we can observe that research in biogas technology showed a sharp increase of articles right before 2010 (Figs. 2, 8, 29, 30 and 31). This exponential increase is consistent with the general increase in scientific publications[18]. Generally, we observe a dominant method inside each field such as sorption methods in biogas cleaning or desulphurisation. The large amount of research on similar methods, might indicate that the field is mature enough to focus more on real world implications such as cost analyses and upscaling as a majority of research is on smaller lab-scale reactors. Unfortunately, this large amount of research is also divided on a large number of different techniques missing some proper standardised definitions and classification.

Firstly, substrate type was collected throughout the different steps reviewed, but this data was unfortunately not exploitable further than for pretreatment reviews. This is due to the very diverse types used and as to the authors knowledge no prior classification of substrate has ever been performed. This is a field that requires increased attention, as there are many parameters to pay attention to such as: water content, solubilisation, Nitrogen and lignocellulose content for example[78]. A proper classification and reporting of substrate might allow future reviews to better identify and reduce unnecessary repetition of experiments.

Pretreatment shows also difficulties with classifications, with different options. In this study physical, thermal and mechanical needed to be included in the same category due to the large scope of the study, using a systematic technique according to measurable definitions, but other different classifications are possible [1,57,122]. Chemical and physical techniques are the main groups with a focus on thermal and sorption techniques respectively. Due to the maturity of this methods, we would recommend focusing more on real world applications than preliminary experiments using these. For chemical techniques, a large number of different compounds were employed to perform similar tasks, a database would prove useful to be able to compare compounds quickly and efficiently, to optimise their use case. Process improvement methods should receive more attention as an emerging branch of pretreatment, due to their positive impact on process stability and methane production, with elements such as biochar showing growing interest [68].

Reactors design showed a clear research focus on high-rate reactors, more focus should be allocated to solid waste, especially with new regulations that make the sorting of food waste compulsory[37]. While the majority of research was performed on small lab scale reactors, a healthy amount of research was observed on larger pilot and full-scale reactor. Nevertheless, there is an issue with clarity and proper word use in this case. The size of the reactors is not always mentioned, and there is no real convention on reactor categories. Pilot scale, for example, is sometimes also used for lab scale reactors regarded as a prototype. A definition issue also arose in the naming scheme with some reactors having multiple names such as filter bed/ fixed bed/ biofilter and the use of the hybrid term for multistage reactors. Such issues can lead to confusion and improper reporting of reactor design and needs to be addressed.

The observation of temperature research showed a specific interest in psychrophilic with a proportionally larger amount of reviews, but this field is poorly studied compared to the mesophilic and thermophilic range. However psychrophilic research should be extended as it shows promise for productivity at lower cost[124]. Research on temperature shows also a very large diversity of combination in multistage reactors and would really benefit from some form of common harmonized reporting to better identify the advantages and disadvantages in different conditions. A better reporting system would also be very advantageous to be able to investigate data by temperature instead of the conventional known terms (psychrophilic, mesophilic and thermophilic). This research would have greatly benefited from it, for more exhaustiveness and objectivity, as some new categories can be missed and research on a finer tune by degree would be very complex.

For the treatment of the gas, both upgrading, and desulphurization use very similar methods and distribution of those. There was a very large number of methods and compounds observed, where a database would prove very beneficial as a traditional paper does not seem suitable for this purpose. A different reporting system would also prove beneficial to observe also different techniques such as mitigation pathways, as those were rejected by the strict definition of a systematic review. Notably, this field is the only one where systematic reviews were observed, with the growing number of scientific articles and the issues of reproducibility increasing, following guidelines, and increasing rigor can be a first step[84]. Other systematic reviews might be present in our data but missed due to human error or a lack of clarity in the text. More problematic, some articles are reported as systematic without any methodology described, as observed for biogas upgrading, making the verification of their results impossible. There is a clear need for systematic reviews in all fields of biogas technology to better understand their strength and weaknesses and compare the methods together.

9. Limitations and prospects

This research showed that there is a need for a modernised reporting system. As this research was not as exhaustive as expected and clear limitations were observed in the traditional reporting system.

This research set up to be exhaustive by using a systematic method on a broad scope, but even in this case many methods were missed. Looking specifically at high-rate reactors, a study review this in detail, not mentioning a systematic review protocol contains examples not seen in this review[120]. Many factors can be responsible both internal and external. The large scope of the work can bring fatigue and increase the chance of mistakes, this is also exacerbated by the lack of standardised reporting and naming conventions. Major external factors include the lack of standardisation in keywords, this can easily be observed. Looking at a random example, searching for "NMMO anaerobic digestion" one of the first examples states the use of NMMO in pretreatment, but this is not visible in the title or the keywords[50]. This issue can also explain why many reactor design were missed as authors often employ specific acronyms "CSTR, UASB,…" without the reactor keyword. More novel techniques without many publications might suffer greatly from this phenomenon where a work can easily be missed if not looking specifically for it. In a world where we are trying to find the best solutions to the current energy crisis it is essential that new methods can easily be picked up. Crucially it shows that a search engine system is also outdated and the use of modern technologies to centralise work and facilitate research is necessary.

Moreover, work could be easily and automatically indexed if standardised reporting was used rendering such a long review procedure unnecessary and greatly reduces the risk of internal identification errors. A classification and reporting method that can be followed by authors could greatly simplify the process and allow for automatic and accurate processes. This would also allow for common definition of concepts, as this was a problem in this study to cover everything accurately and the avoidance of the need to redefine every concept if a standard library could be found. This would also allow for quick comparison between techniques, as this is one crucial part missing for this work, due to the scope, the time and effort required for comparing methods depending on the different use cases this would not be feasible, but a standard reference library would allow for readers to quickly identify where to find such information and for quicker cooperation to fill the gaps needed by researchers.

This work also highlighted the issues with open science and reproducibility in this field. Production of clear guidelines for reporting and comparison and cooperation in the field could help solve the current "reproducibility crisis". There is a lack of systematic review work, with the growing number of scientific articles and the issues of reproducibility increasing, following guidelines and increasing rigor can be a first step[84]. Other systematic reviews might be present in our data but missed due to human error or a lack of clarity in the text. More problematic, some articles are reported as systematic without any methodology described, as observed for biogas upgrading, making the verification of their results impossible. There is a clear need for systematic reviews in all fields of biogas technology to better understand their strength and weaknesses and compare the methods together. Another issue that emerged at every step was the lack of access to articles, which forced the exclusion of otherwise valid articles. Outside the scope of this research, the lack of standardisation on performance evaluation to better compare results is also an example of this type of issue [14].

A traditional paper format does not seem to be able to fit such review work as it has a rigid and not updatable structure. Comprehensive reviews such as covering many digesters [120], can quickly become outdated with new data on specific digesters and new type of digesters. As the growth of scientific literature is exponential[18]and this field shows a similar pattern. There is an urgent need to find modern solutions to this issue and deal with this reality. A solution could come from new technologies such as "living papers" or databases that can be updated over time. Extensive work like this one, could be extended by more research and facilitate data verification and comparison. Other branches such as biomedical science have already started using this model with great results and faster progress[108]. This system could also ensure to include all areas of research as the rigidity of a systematic review cannot include everything as mentioned before. Moreover, some important fields might be missed, in this context the digestate treatment was not reviewed due to constraints, although its management is increasingly an issue. Any technology patented or used in the industry might not necessarily be mentioned in such a systematic review but could be easily added to a cooperative living work. Lastly, such a system would allow for quickly identify and request research in specific areas such as explaining the advantages or disadvantages and comparing work. For example, it is not always clear why certain reactors are more used than others. More cooperation and centralisation needs to take place for a system like this to work, it could help provide clear guidelines for reporting and the gathering of information could help automate and increase review work quality, while making it easier for researchers to report and now what to report.

10. Conclusion

This work covered 4660 articles over 5 major steps of biogas production and highlighted progress and new techniques used. The field is relatively new with a general increase in research around 2010, with a well-balanced research focus on the 5 fields. There are presence of dominant methods, in each field with usually a lot of minor researched methods. However, some challenges remained due to a lack of standardisation in the scientific work. This leads to tedious work to recover information and the inevitability of missing crucial information. This lack of standardisation makes identifying and comparing different methods complex, as the field is growing quickly with a large variety of different methods employed in different scenarios.

Even with specific effort in exhaustiveness and systematic process, this work failed to be exhaustive, and many interesting works were missed, due to multiple factors. This work however, showed the limit of traditional reporting. There is a clear need of modernised techniques in reporting science, as many issues in classification, definitions, standardisation and reporting could be tackled by more cooperative and updatable work, not hampered by the rigid paper system, not adapted to such work. Moreover, instead of a tedious long process with possible errors, such work could be automated in a modernised system using clear standards and modern systems such as databases. Modernisation is necessary to ensure faster information transfer and progress.

Lastly, it was also highlighted that this field also need an open science framework system promoting, reporting guidelines, cooperation and rigour, as there is still a lack of systematic review and accessibility to content can be an issue.

CRediT authorship contribution statement

Antoine Bercy: Writing – review & editing, Writing – original draft, Visualization, Validation, Methodology, Investigation, Formal analysis, Data curation. **Hynek Roubík:** Writing – review & editing, Validation, Supervision, Project administration, Methodology, Conceptualization.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

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