



Review article

Approaches to mitigation of hydrogen sulfide during anaerobic digestion process – A review

Eric Mutegoa^{*}, Mtabazi G. Sahini*Department of Chemistry, College of Natural and Mathematical Sciences (CNMS), The University of Dodoma, P.O. Box 338, Dodoma, Tanzania*

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ABSTRACT

Anaerobic digestion (AD) is the primary technology for energy production from wet biomass under a limited oxygen supply. Various wastes rich in organic content have been renowned for enhancing the process of biogas production. However, several other intermediate unwanted products such as hydrogen sulfide, ammonia, carbon dioxide, siloxanes and halogens have been generated during the process, which tends to lower the quality and quantity of the harvested biogas. The removal of hydrogen sulfide from wastewater, a potential substrate for anaerobic digestion, using various technologies is covered in this study. It is recommended that microaeration would increase the higher removal efficiency of hydrogen sulfide based on a number of benefits for the specific method. The process is primarily accomplished by dosing smaller amounts of oxygen in the digester, which increases the system's oxidizing capacity by rendering the sulfate reducing bacteria responsible for converting sulfate ions to hydrogen sulfide inactive. This paper reviews physicochemical and biological methods that have been in place to eliminate the effects of hydrogen sulfide from wastewater treated anaerobically and future direction to remove hydrogen sulfide from biogas produced.

1. Introduction

Anaerobic digestion (AD) is a microbiological process widely used to produce biogas. This process is carried out under anaerobic conditions using organic waste as a raw material or substrate for anaerobic microorganisms [1]. Generally, biogas is a mixture of methane and carbon dioxide, the methane content being in the range of 50–70% [2]. Recently, the world has been turning from utilizing fossil fuels as an energy source for different purposes such as cooking, lighting and machinery operation [3,4]. Due to the increase in the costs of oil and natural gas on the global market as a result of fossil fuel depletion, there is a considerable increase in the demand for other alternative energy sources [5,6]. However, owing to the advancement of science and technology, it is recommended that other alternative sources of energy should be used as a direct replacement for these commonly used fossil fuels, which are not environmentally friendly considering the release of large amounts of carbon dioxide in the atmosphere when burnt.

Furthermore, the use of these alternative sources of energy is cost-effective due to sustainability and its contribution to clean energy generation and protecting the environment against global warming [7,8,9]. In order to provide alternate energy sources with little sludge generation, anaerobic digesters have been used all over the world. High-rate anaerobic digesters are commonly used in the treatment of sewage. This is due to the fact that high-rate anaerobic reactors, as opposed to conventional low-rate anaerobic digesters, are designed to operate at short hydraulic retention times (HRT) with higher solid retention times that can accommodate large

^{*} Corresponding author.

E-mail addresses: eric.mutegoa@gmail.com, eric.goshoko@udom.ac.tz (E. Mutegoa).

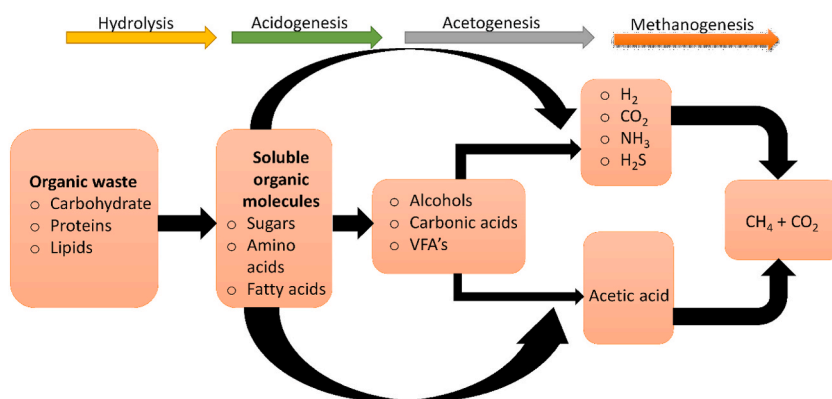


Fig. 1. Anaerobic digestion process of organic waste.

amounts of biomass at once, improving sludge stabilization and increasing loading capacity [10]. The Upflow Anaerobic Sludge Blanket (UASB) reactor is one of the most widely utilized high rate digesters due to its effectiveness in removing organic matter and the high-quality amount of biogas it produces [11].

Biogas technology is among the ecofriendly processes in producing methane gas from various organic substrates such as cow dung [12,13], pig manure [14,15], industrial effluents [16,17], food waste [18], slaughterhouse waste [19,20] and agro-waste [21,22]. These wastes are plentiful and cheap to acquire since they are being discharged to the environment as a result of both domestic and industrial activities. However, the production of biogas is sometimes associated with other contaminants such as siloxanes [23], total ammonia nitrogen (TAN) [24], hydrogen sulfide [25,26], halogens [27,28] and other volatile organic compounds [29,30] which becomes inhibitory to methanogens and subjecting the whole digestion process to failure. Hydrogen sulfide (H_2S) is mainly produced as an intermediate product during the AD process, in which excessive production significantly impacts the final stage of methanogenesis [31]. The produced H_2S during acetogenesis tends to affect the growth of methanogens, which are required to initiate the methanogenesis process, as indicated in Fig. 1.

In most cases, biogas production during anaerobic digestion of carbon-based substrate is highly influenced by sulfide deposits in the digester resulting from the protein breakdown process [32,33]. Generally, substrates rich in microbial sulfate and other sulfur-containing compounds are subjected to the release of hydrogen sulfide, which is noxious to humans and corrosive to various appliances and metal pipes, when treated both aerobically and anaerobically [34,35]. The aftermath of sulfide inhibition, which induces the antagonism between sulfate-reducing bacteria (SRB) and methane-producing archaea (MPA) throughout the AD process [36,37], releases hydrogen sulfide with a bad odour to the environment. Furthermore, when mixed in a higher amount with the biogas, the gas tends to corrode the surfaces of the cooking devices and other equipment utilizing biogas as fuel, for example, generator sets and several other automobile engines [38,39]. H_2S can result in various specialized damages, such as corrosion-mechanical attacks, tool and equipment failures, and difficulties with tool joints (Table 4). Regardless of how the biogas will be used, hydrogen sulfide must be removed to variable degrees because its presence increases the cost of operations and maintenance and shortens the lifespan of gas handling equipment. Various studies have reported that SRB's effects on anaerobic processes go beyond reducing methane production due to consuming organic waste and creating hydrogen sulfide. For instance, Uberoi and Bhattacharya's research found that as the metabolic rate dropped with increased COD load, the ratio of chemical oxygen demand (COD) to sulfate (SO_4^{2-}) had an impact on the SRB's effect on anaerobic digestion [40].

In practice, the resiliency of the acetoclastic methanogenesis pathway is crucial for effective methane synthesis during the switch from acidogenesis to methanogenesis. The initial stage of anaerobic digestion, known as acidogenesis (Fig. 1), involves the conversion of complex chemical compounds into simpler molecules such as acetate and volatile fatty acids [46]. These intermediary substances are transformed into methane during the process of methanogenesis, which comes after acetogenesis. Several factors, including the effects of mechanically mixing or agitating the anaerobic digestion system, can affect the stability of the acetoclastic methanogenesis pathway during this transition [47].

The ideal agitation conditions for the stability of the acetoclastic methanogenesis pathway may differ depending on the particular reactor design, feedstock properties, and the microbial population present [48]. To ensure the stability and effectiveness of the methanogenic process, rigorous monitoring and control of agitation parameters, such as mixing intensity and duration, is required.

On the other hand, temperature optimization is essential to produce biogas effectively in mesophilic AD. The activity of hydrolases and microbial metabolism can be increased by higher temperatures, hastening the breakdown of organic waste and increasing biogas output [12,49]. On the other hand, extremely high temperatures might stifle microbial activity and enzyme activity, resulting in unstable processes and reduced biogas generation [20].

Therefore, evaluating which method fits best in mitigating hydrogen sulfide is paramount during anaerobic digestion to produce high quality biogas.

This study has meticulously evaluated several approaches for easing the inhibitory process caused by sulfide levels forming in the digester and divided into two categories: physicochemical and biological methods.

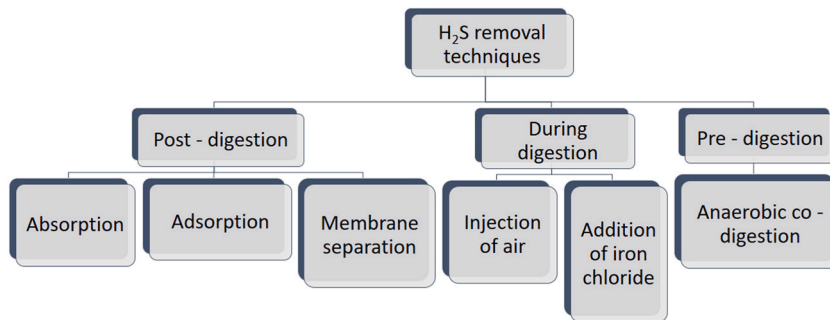


Fig. 2. A summary of different techniques for H₂S removal [50].

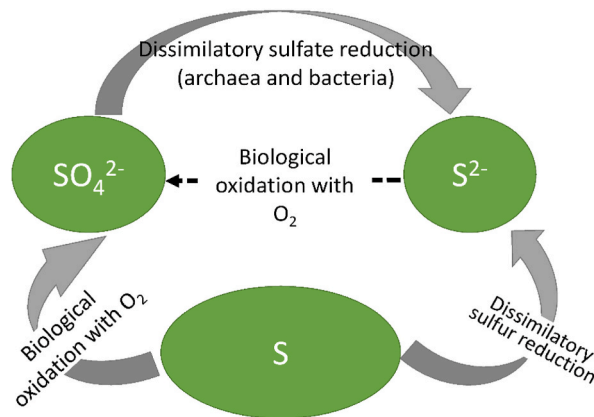


Fig. 3. The cycle of microbial sulfur during anaerobic digestion [59].

2. Physicochemical procedures

The physicochemical method for eliminating hydrogen sulfide from the biogas is a straightforward process that occurs in the reactor through diverse ways in which sulfide oxidizing bacteria is treated as a potential additive being added to oxidize sulfide to elemental sulfur. The post-digestion process in physicochemical procedures, however, involves using a variety of absorbents and adsorbents as a means of post-treatment, including activated carbon and wet scrubbers (Fig. 2). These materials directly remove hydrogen sulfide (H₂S) through sorption interactions with the biogas.

This procedure primarily uses the micro-aerobic process, precipitation, and scrubbing. However, biological desulfurization technology is a long-term method for removing sulfide from biogas. Its benefits include minimal chemical agent usage, little surplus sludge production, and effective elemental sulfur recycling. Hydrogen sulfide is initially transported to the liquid phase by the alkaline absorption solution in the current sulfide removal systems (Fig. 4), where sulfur-oxidizing bacteria (SOB) then oxidize it to sulfur or sulfate, resulting in a high sulfide removal efficiency [51].

2.1. Micro-aerobic process

This method has been extensively used (as shown in Fig. 2 during air injection) to remove hydrogen sulfide from biogas because it is efficient, simple, and cost-effective [52]. The addition of air into an anaerobic digester is used in this technique [53]. The method enhances sulfide oxidation by introducing oxygen in the reactor containing sludge under micro-aerobic conditions [54]. This method uses oxygen as an electron acceptor during sulfide oxidation, in which excess sulfide is further transformed into elemental sulfur, as shown in equation (1).

Essentially, when sulfide is oxidized to elemental sulfur (S) or sulfate (SO₄²⁻) forms, the process is regarded as the basic principle for hydrogen sulfide elimination [55]. Meanwhile, thiosulfate (S₂O₃²⁻) may be produced during the process, with sulfide as the terminal electron acceptor throughout [56]. Sulfur is the primary end-product of sulfide oxidation under a limited supply of oxygen (micro-aerobic) conditions, especially when the oxygen level is less than 0.1 mg/L, as shown in the equation below:



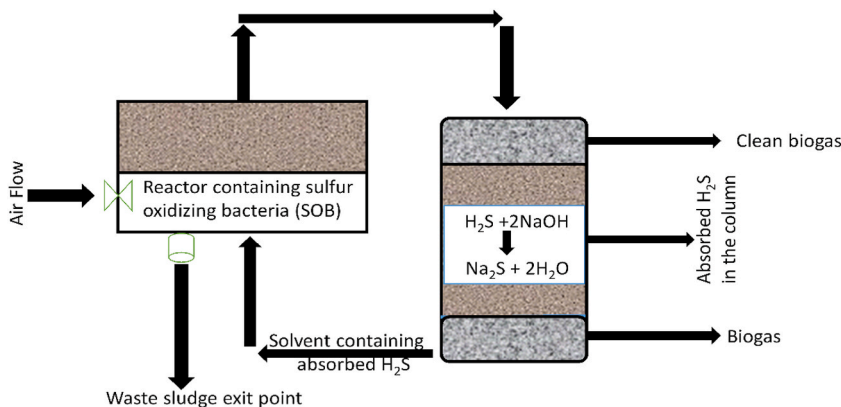


Fig. 4. Biological desulfurization process modified from reference (Okoro and Sun, 2019).

Table 1

The impact of inoculum to substrate ratio on methane yield for selected substrates.

S/N	Substrate	Inoculum	Inoculum to substrate ratio (ISR)	Methane yield (ml CH ₄ /g VS added)	Average number of days	Reference
1	Ball milled straw (BMS)	Cattle manure	10	<50	150	[70]
			30	>200		
			70	>300		
2	Kitchen waste	Industrial sludge	0.5	220	25	[71]
			1.0	180		
			1.35	200		
			2.3	220		
			3	7023		
3	Maize	Digester sludge from a municipal wastewater treatment plant	2	11,356	20	[72]
			1.5	14,921		
			1	23,635		
			1	23,635		

In general, controlling the production of sulfur and sulfate depends on the quantity of oxygen supplied in the reactor (Fig. 3) for the sulfide oxidation process in which elemental sulfur is regarded as an intermediate product undergoing disproportionation [57,58]. For example, 0.5 mol O₂/mol S²⁻ is thought to be necessary for the oxidation process of sulfide to elemental sulfur (Eqn. (1)). Nevertheless, 2 mol O₂/mol S²⁻ is considered to be required for the oxidation process of sulfide to sulfate (Eqn. (2)). The formation of sulfate depends on the limited conditions of sulfide in which the higher amount of oxygen consumed per 1 mol of sulfide will enhance sulfate formation [55].



Meanwhile, Jung and Mora, in their study [60,61], reported that at oxygen levels more than 25 μmol O₂/0.04 mol S²⁻, sulfide was predominantly absorbed and oxidized to elemental sulfur. This trend is similar to the findings of Tilahun and fellow researchers [62], who discovered that dissolved oxygen (DO) concentrations significantly impacted the formation of S⁰. However, there are additional important aspects to consider for an operative micro-aeration approach, such as oxygen transfer rate (OTR) and oxygen utilization rate (OUR), in addition to the quantity of oxygen prerequisite to augment the micro aeration course [63]. The two processes are considered the limiting components since they are essential in feeding the reactor sufficient oxygen for sulfide removal. Reactor conformation, micro-aeration method (i.e., usage of air or oxygen, bubble size, injection in aqueous or gaseous phase), Total solids (TS) concentration of substrate in the reactor, and other parameters all influence the OTR throughout the AD process [64]. Once the dispersion of air/oxygen in the reactor was inadequate, for example, the micro-aeration process failed to eliminate sulfide in the liquid phase [49]. On the other hand, the inoculum deployed and the substrate injected into the reactor during the AD process affects the OUR in micro-aeration. Initially, it was revealed that the microaeration process requires a particular oxygen dosing method for substrates with an increasing rate of hydrolysis, such as lignocellulosic biomass [63].

Meanwhile, the conversion of organic materials such as agricultural residues, municipal solid waste, abattoir waste, and others into methane has been fueled partly by the inoculum to substrate ratio (ISR) [65]. The optimal ISR in the reactor enables the presence of responsible bacteria for each stage involved during the AD process [66]. Initially, the inoculum used for optimal biomethane conversion has a number of characteristics, such as fewer pollutants from external sources that could interfere with the entire process. To ensure that the microbial species are acclimated, it is suggested that the inoculum be similar to the substrate being fed into the reactor. However, to attain higher levels of methane yield, the inoculum should be fresh and include the minimum viable inhibitors, such as hydrogen sulfide, ammonia, and heavy metals [67]. Generally, the highest ISR level often ensures a larger methane yield because it

inhibits the acidification process and maintains the nutritional balance for microbes [68]. Nevertheless, as the inocula dosage increases, the reactor's performance improves (Table 1). By optimizing these factors, it is possible to enhance the efficiency of the AD process and generate higher methane content yields [69].

2.2. Precipitation process

The precipitation method, which employs metals with a strong affinity for sulfur, such as iron and zinc, has been one of the most common techniques for treating sulfur-containing wastewater. Metal salts such as iron, zinc, lead, and copper are added to precipitate sulfide. For instance, adding iron chloride during digestion (Fig. 2) results in the formation of highly insoluble metallic sulfide precipitates [73,74]. The metal sulfide plays a major role in reducing the solubility product and lowering sludge volume compared to hydroxide precipitation. However, sulfide precipitation is preferred over hydroxide precipitation because of its low solubility and lower sensitivity to pH changes. Because the solubility of metal sulfides is not as sensitive to pH changes as that of metal hydroxides, in addition, it is easier to recover metal from metal sulfides than hydroxides since sulfide precipitation results in lower effluent concentrations and less interference from chelating agents [75].

When treated with metal-containing wastewater, sulfate-reducing bacteria (SRB) display potential application over other chemical treatments. In the liquid phase, Fe^{3+} is quite effective at reducing sulfide. While being reduced into Fe^{2+} , Fe^{3+} oxidizes sulfide to elemental sulfur, which precipitates with sulfide to generate ferrous sulfide precipitants [76,77,78].



For example, the study which was done by Zhao and other co-authors [79] discovered that Ferric iron (Eqn. (3)) is normally used for sulfide precipitation in sewers, thus conquering corrosion and odour control. According to the study, monitoring the sulfide level during precipitation enhances the evaluation of the performance of sulfate-reducing bacteria and methanogens in anaerobic sewer biofilms. Sewer biofilms were shown to be severely inhibited in reducing sulfate and producing methane when Fe^{3+} was added during sulfur precipitation in the liquid phase. These inhibiting effects are caused by the development of iron sulfide (FeS) precipitates (Eqn. (4)) and modifications to the dynamics of microbial communities, which an optimized dosage of Fe^{3+} addition can minimize.

However, sewer biofilms are affected by several factors, including pH elevation, which can promote the growth of some bacteria while inhibiting the growth of others, thereby causing changes in the composition of the microbial community [80].

On the other hand, time-based succession, which describes the slow changes in microbial populations over time, occurs in biofilms in sewer systems. As a result of factors like nutrition availability, organic substrate composition, and ambient circumstances, various microbial communities flourish at various phases.

In addition, by adjusting the sulfur cycle processes in the biofilm, ferric iron can help reduce sulfide concentrations and manage odours [81]. This is because some microorganisms engaged in sulfur metabolism can use ferric iron as an electron acceptor. It could foster the growth of sulfur-oxidizing bacteria, which use ferric iron as a source of energy to convert sulfide to sulfate.

Meanwhile, due to the precipitation process generated by the Fe^{3+} dose, the effluent emitted from the reactor produced a more significant amount of sulfate at the end of the process. The sulfide concentration in the reactor was reduced by about 60%, indicating that consistent Fe^{3+} addition would be considered a viable solution to lower the inhibition process due to sulfide. In other situations, it has also been ascertained that ferric iron has little effect on the precipitation reaction in acidic conditions. For instance, the sulfide elimination process to the level of 60% can be attained at a reduced time proportion of 1.5 s when the pH for wastewater is set at 7 [82]. The analysis of the significance of pH through precipitation reaction displays that the acidic condition can induce sulfide precipitation when the excess ferrous iron of about 40% is used, according to the study. At an alkaline medium ($\text{pH} > 8$), however, the result reveals that all of the additional iron (II) salts can be precipitated completely [83]. A 1:1 ratio of iron (III) to iron (II) should be perfected by mixing ferric chloride and ferrous sulfate to maximize the efficacy of the precipitation reaction. This is due to the fact that a combination of Fe(II) and Fe(III) has been proven to improve the recovery process of sulfide from wastewater rather than relying on either salt alone [73].

However, aromatic and sulfur functional groups in wastewater can effectively increase the precipitation process of metal sulfide by providing additional sites for metals to attach to sulfide [84]. In practice, the delocalized electrons in benzene rings and other aromatic compounds stabilize the bonding process with metal ions [85]. As a result, these metal ions form strong complexes with functional groups containing sulfur and sulfide. Since the generated metal-sulfur interactions are often insoluble, they can additionally be precipitated during the wastewater treatment process and serve to lower the sulfide concentration in wastewater.

Therefore, the augmentation of aromatic and sulfur functional groups in wastewater plays a crucial role in enhancing the affinity of metal – sulfur interaction, forming an insoluble curdle in the reactor. Sulfur can be removed from wastewater more effectively by precipitating out the insoluble metal complex that forms when metal ions like zinc or iron bind with sulfur.

Nevertheless, the addition of distinctive coagulants such as partial precipitants of ferric chloride and ferrous sulfate together with the hydroxides and carbonates of calcium in this manner being treated as coagulant-aids enriched the higher sulfide removal effectiveness in the range of 96–99% when the physicochemical technique was employed [86].

2.3. Adsorption of hydrogen sulfide (H_2S) on activated carbon

Adsorption is a regular process for pollutant removal using physical and chemical procedures to eliminate adsorbed material in the form of adsorbate [87,88]. Adsorption is the process through which a material (adsorbate or sorbate) builds up on the surface of a solid (adsorbent or sorbent) [89]. The adsorbate might exist in a liquid or gaseous phase. Unsaturated forces at the solid surface that have the potential to form bonds with the adsorbate are the driving force behind adsorption [90]. For example, when iron metal is impregnated with an adsorbent such as biochar, H_2S removal efficiency is enhanced from biogas using a biogas scrubbing column. The reactive oxygen and metal oxides in biochar can favour the conversion of sulfide into elemental sulfur and sulfates, which will further increase the H_2S removal capacity [91]. During the AD process, this phenomenon is done after breaking down the biomass into biogas, as shown in Fig. 2 (Post digestion).

Meanwhile, Nguyen-Thanh and Badosz investigated the effects of bentonite clay binders containing copper, zinc, or iron in the interlayer gaps on the adsorbents' ability to remove H_2S . The adsorbent capacity of AC was then reported to be improved by surface modification with a binder containing copper, indicating how the oxygenated surface groups were aiding the adsorption process. For instance, an increase in surface acidity of about half a pH unit was found for carbon with binders [92]. Additionally, it has been investigated whether adding phosphates during AD can increase the effectiveness of H_2S elimination. Trisodium phosphate (TSP) and monosodium phosphate (MSP) are examples of phosphates that can react with H_2S to produce precipitates of insoluble metal sulfide [93]. Compared to gaseous H_2S , these precipitates are easier to capture or remove from the gas phase. Phosphates can lower the potential of H_2S release into the biogas and enhance overall H_2S removal effectiveness.

The study by Ansari and others [94] demonstrated the importance of composite materials mixed in various amounts, which examined the impact of the concerted outcome of the modified surface properties of sewage sludge attained from materials bestowed with carbon. Compared to the adsorbents generated from pure precursors, the findings demonstrated that combining the polymer with sludge enhances the adsorption of hydrogen sulfide by 50%. The material's improved performance is due to the oxidation process of hydrogen sulfide influenced by catalytic centres from the sewage sludge. At the same time, a carbonaceous phase depicts a key role in boosting catalytic centre distribution by providing greater space for storage in its micropores. Due to the increased surface area and chemical interactions between the polymer and the sludge, this combination can be successful at adsorbing various sulfur-containing compounds, such as polysulfides and selenyl sulfides, despite the fact that its main focus is frequently on hydrogen sulfide (H_2S) adsorption [95].

Conversely, sludge with a variety of binding sites and polymers with a high affinity for sulfur compounds are able to capture and immobilize these pollutants, lowering their concentration in the treated media, similarly to the adsorption of H_2S . However, it's important to understand that the sorbent's precise design and functionalization methods will depend on the intended application, the H_2S concentration, and the operating circumstances. Extensive testing and optimization studies are necessary to achieve the desired performance in removing H_2S .

2.4. Stripping method

The technology is useful since it enhances energy recovery through biogas during the AD process while producing minimal sludge [96]. This method entails the creation of a stripper during anaerobic digestion capable of extracting sulfide from wastewater while maintaining the effluent's chemical properties. For example, an inventive stripper constructed in the laboratory with regulated factors encompassed during the design and set-up, such as airflow rate, liquid flow rate, liquid-to-air ratio, and pH profile, was claimed to have a 60–70% sulfide removal efficacy. The design imposed the stripper from the bottom, and the sulfide wastewater was passed from the top [97]. During stripper designing, adjusting the incoming air entailed per amount of sulfide in the particular experiment is critical. The amount of air fed into the stripper is one of the critical design control points. When the air is not properly controlled, sulfur dioxide (SO_2) can be produced through hydrogen sulfide (H_2S) oxidation. In addition, since it impacts the reaction kinetics of H_2S oxidation, the temperature of the stripper needs to be carefully controlled [98]. However, to ensure sufficient contact between H_2S and the oxidizing agent, it is also necessary to monitor the residence time of the gas stream inside the stripper. Controlling these critical components is essential for minimizing the risks that could result from poor designing of the stripper. Prioritizing risks include H_2S release, SO_2 generation and corrosion [99].

Before being released into the atmosphere, hydrogen sulfide-contaminated air must be cleaned using an essential tool such as a biofilter [100]. Initially, the process relied on air recycling, with recycled air emitting traces of hydrogen sulfide into the atmosphere. Recycling stripper wastewater, which may include a lower percentage of free H_2S , might be considered a sulfide concentration control strategy in the anaerobic reactor. When monitoring low concentrations of H_2S , passive samplers consisting of polyethylene adsorbing cartridges loaded with zinc acetate are considered simple, portable, and economical to use [101]. However, it has been reported that air recycling and air injection into wastewater effectively reduce H_2S [102]. The process of pumping air into the reactor often raises the dissolved oxygen concentration, favouring the aerobic process and reducing the growth of the anaerobic bacteria that produce H_2S . On the other hand, recycling the air in the reactor under limited pressure substantially lowers the release of H_2S by improving the treatment systems through optimization of the aeration system efficiency.

2.5. Wet scrubbing

Wet or chemical scrubbing or absorption involves the mass transfer of H_2S in the liquid phase aided by acidic, basic, or oxidant reagents. The sodium hypochlorite solution ($NaClO$) oxidizes hydrogen sulfide (H_2S) to sodium sulfate (Na_2SO_4) in the alkaline media

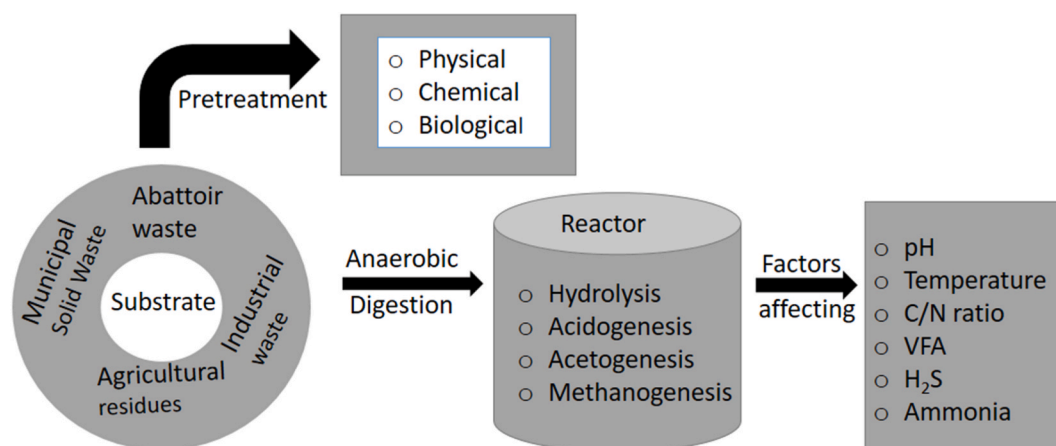


Fig. 5. Factors affecting microbial hydrolysis during the anaerobic digestion process.

(pH > 9), as indicated in the equation below (Eqn. (5))[103].



From the study done by Biard et al. [103] on the enforcement of the scrubber with a contactor where solvent and gas flow co-currently is at a high velocity greater than 12 m s^{-1} , it was reported that hydrogen sulfide suppression efficacy of 95% could be achieved. This high removal efficiency is complemented by the residence time of the solvent in the scrubber being shortened to 30 m s (microsecond) using sodium hypochlorite (NaOCl) solution. This study used a single factor research approach for process parameter optimization, which meant that while one process parameter or factor was examined, all other parameters or factors remained unchanged [104]. According to these findings, the maximum H₂S removal effectiveness was 98.2% under ideal experimental circumstances. Furthermore, it was reported that as UV-light intensity and solution pH were improved, the H₂S removal efficiency increased. In this experiment, whereby hydrogen peroxide (H₂O₂) was employed as one of the oxidizing agents and urea as an alkaline media, the change of the H₂O₂ concentration from 0 to 0.2 mol/L stimulated the rise in H₂S removal efficiency firstly at 38.7%–98.2% and then lessened to 98.2%–90.1%. Because of the high solubility of sulfur dioxide (SO₂) in water, various studies indicate that the removal effectiveness of SO₂ in the wet-scrubbing method was about 100% [105,106,107,108]. Furthermore, this method is extensively utilized due to its ease of use and great efficiency. However, using various absorbers necessitates a high regeneration energy input and non-corrosive equipment [109,110]. On the other hand, chemical scrubbing in packed towers may entail some running costs, as it necessitates building high and big wet scrubbers [111].

3. Biological methods

Biological desulfurization technology is a long-term method for removing sulfide from biogas. Its benefits include minimal chemical agent usage, little surplus sludge production, and effective elemental sulfur recycling. Hydrogen sulfide is initially transported to the liquid phase by the alkaline absorption solution in the current sulfide removal systems (Fig. 4), where sulfur-oxidizing bacteria (SOB) then oxidize it to sulfur or sulfate, resulting in a high sulfide removal efficiency [51].

In order to evacuate hydrogen sulfide from wastewater treated for the AD process, physicochemical processes such as absorption have been used [112,113], scrubbing [114], and using water-containing chemicals [115,116]. Nonetheless, these procedures are expensive and can result in chemical waste accumulation. Biological treatment procedures such as autotrophic denitrification [117] and biological desulfurization [118], on the other hand, have been preferred over other techniques due to the method's moderate operating conditions and low cost.

Two major techniques under this method are involved during the process; aerobic and anaerobic (or anoxic), depending on various electron acceptors. The electron acceptor in the aerobic technique is usually oxygen, whereas, in an anoxic approach, the electron acceptor is often nitrate or nitrite [119].

3.1. Aerobic biological technology

The microaerobic process involves using a low concentration of dissolved oxygen (DO) for microorganisms to facilitate hydrolysis acidification. However, the aerobic process is referred to as extended aeration, in which plenty of aeration is needed for energy derivation from carbohydrates (sugars). In practice, the microbes will start feeding on each other under the deprived aeration process [120]. Through enzymatic processes, microbial hydrolysis transforms complex chemicals into volatile fatty acids that microbes can use in subsequent stages of the AD process [46]. In order to produce an environment suitable for various microbial activities, the level of DO is often kept low (Fig. 4). However, several factors, including temperature, pH, nutrient availability, and substrate characteristics,

Table 2

A summary of the strategies used to control sulfide inhibition by the AD system and the conditions under which they were employed.

Method/ Technique	Working Principle	Appropriate condition(s)	Reference
Micro-aerobic process	The method uses oxygen as an electron acceptor during sulfide oxidation, in which excess sulfide is further transformed into elemental sulfur.	<ul style="list-style-type: none"> - Temperature reactor of 30 ± 1 °C - Adoption of wastewater with a higher carbon to nitrogen (C/N) ratio (1.26:1) for stimulating the growth of heterotrophic bacteria. - Aeration rate of 20 mL/min for enhanced sulfide removal - Optimal molar ratio of 0.5–1 for O_2/S^{2-} 	[124,63, 125,52]
Precipitation process	The method employs metals with a strong affinity for sulfur, such as iron and zinc.	<ul style="list-style-type: none"> - Optimum pH value (4.61–6.95) - Low-voltage electrochemical treatment (0.7–1 V) to avoid oxygen generation during anaerobic digestion. - High range of temperature (300–800 °C) for metal oxide precipitating with H_2S. 	[126,127, 128]
Adsorption process	A material (adsorbate or sorbate) builds up on the surface of a solid (adsorbent or sorbent).	<ul style="list-style-type: none"> - High surface area of the adsorbent (500 m^2/g) - Chemically activated adsorbent - Optimal calcination temperature of 800 °C - Pretreatment methods - Nitrogen, oxygen, and sulfur functional groups integrated with the carbon structure 	[129,130, 131]
Stripping	A physical system in which air and wastewater flow in opposite directions under mass transfer.	<ul style="list-style-type: none"> - High operating temperature (above 45 °C) - Stripping tower for volatilization of H_2S 	[132,133]
Wet scrubbing	The method uses water as an adsorbent since H_2S is more soluble in water than methane.	<ul style="list-style-type: none"> - Higher Ultra Violet (UV) light intensity of about 132 $\mu W/cm^2$ - Solution pH of 7.61 - Gas flow rate is about 1.5 L/min - H_2O_2 concentration of 0.2 mol/L - Low temperature (10 °C) - Clean air with little traces of H_2S is discharged from the reactor and can be discharged into the atmosphere 	[134,135]
Aerobic biological technology	The process involves using a low concentration of (DO) for microorganisms to facilitate hydrolysis acidification.	<ul style="list-style-type: none"> - Gas inlet load over 55 g $S-H_2S m^{-3}h^{-1}$ - 1:1 M ratio of N/S - Hydraulic residence time (HRT) of 36–40 h. - Gas residence time (GRT) of 41s. - Optimal pH range of 7–8. 	[136,137, 60]
Biofiltration	Contaminated gases are carried into the biofilm and through the biofilter, where microbes use them as carbon or energy source. The H_2S is further broken down to produce sulfate or sulfuric acid.	<ul style="list-style-type: none"> - Maintaining a low level of oxygen, $\leq 5\%$. - Low pH condition of 2–3 - Microbial community inoculation for converting H_2S to SO_4^{2-} or S^0 	[138,139, 140]

impact the hydrolysis process [121]. Different hydrolysis settings are preferable depending on the particular microorganisms involved and the nature of the substrate being used. The control of the acidification process in the reactor depends critically on optimizing these factors for the hydrolysis progression (Fig. 5).

The quantity of air flow or (DO) in the reactor comprising wastewater for the removal of sulfide was examined in various experiments with this approach. For example, in their study, Guerrero and other co-authors [122] ascertained that the ratio of oxygen to sulfide, determined by the chemical reactions below (Eqns. (6)–(8)), affects sulfide removal effectiveness under aerobic conditions.:



This method revealed that a maximum of 90% sulfide removal could be attained at optimal dissolved oxygen levels. The findings show that the minimum amount of oxygen can produce the highest sulfide removal efficiency. These results agree with the study of other scholars, such as Doğan and coworkers [123]. They run biological sulfide oxidation by operating an airlift reactor under oxygen-limited conditions (0.2–1.0 mg/L), as shown in Table 2. It was discovered in this investigation that as the volumetric sulfide loading rate was improved, elemental sulfur generation rose as well, with sulfide removal exceeding 93%.

The mass transfer of H_2S molecules during removal can be effectively controlled by adjusting the pressure, airflow rate, and silicone membrane design [141]. However, silicone membranes and silicon carbide nanocages utilized in gas sensing have multiple applications and work based on various mechanisms. While silicon membranes use selective permeability to separate gases, silicon carbide nanocages use adsorption or interaction with gas molecules as the basis for operation [142]. Generally, silicone membranes are

Biological Desulfurization under Anoxic Conditions

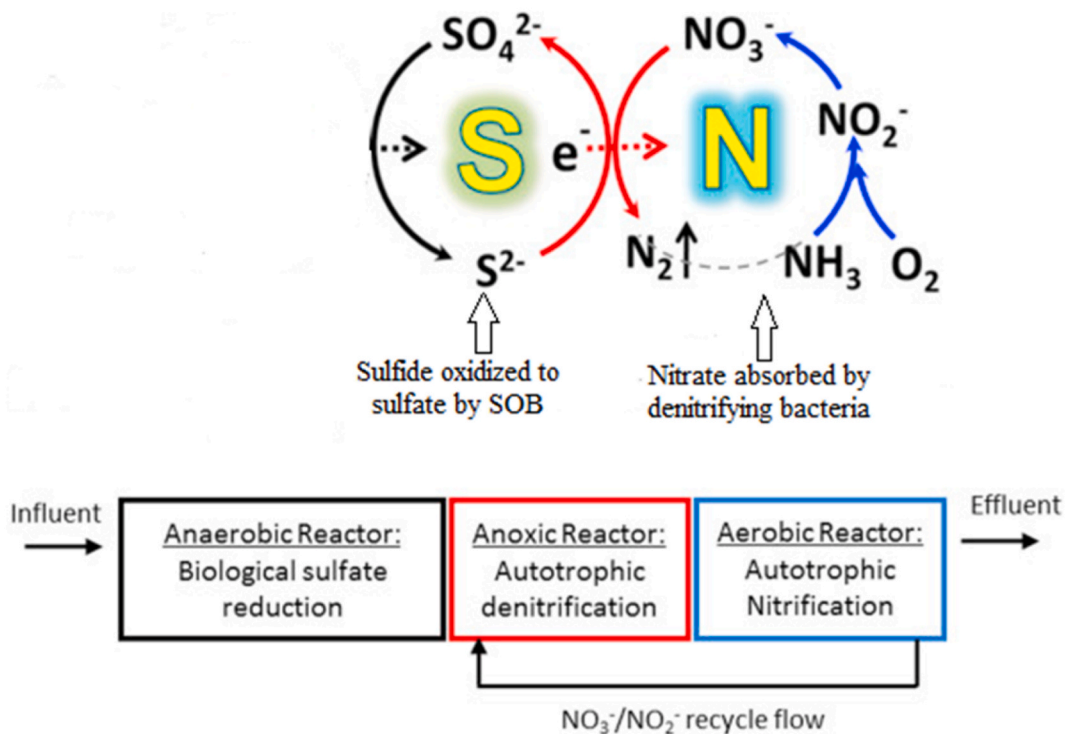


Fig. 6. Biological (Anoxic) Desulfurization Process modified from reference [150].

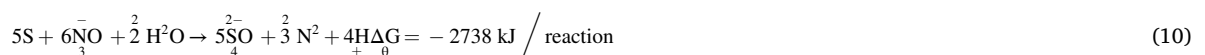
designed to provide high separation efficiency and eliminate particular gases from gas streams, such as H_2S [143]. On the other hand, the detection and measurement of gas molecule concentrations, including H_2S , are the main objectives of silicon carbide nanocages [144]. Both have distinct capabilities that suit them for particular gas treatment and analysis applications.

In a combined anaerobic/microaerobic reactor with no significant sulfate production, this technique increased sulfide removal efficiency to around 96% [119].

However, biogas to air ratio increases and retention time (RT) were crucial features analyzed and proposed to influence sulfide removal efficiency. It was discovered that RT and increasing air mix ratio improved H_2S removal efficiency. According to Chaiprapat and co-authors' research, the biogas-to-air ratio of 1:4 was adequate for H_2S removal, with average reductions of 94.7%, 87.3%, and 85.6% for the biofiltration system reactor at retention times of 160, 80, and 40 s, respectively [145].

3.2. Anaerobic (anoxic) biological technology

The anoxic method describes a route in which denitrifying bacteria absorb nitrate to remove sulfide through sulfide oxidation (Fig. 6). Because nitrite and nitrate concentrations in wastewater are small, these quantities are intentionally fed into the digester during the sulfide removal process [146]. Even though the nitrate-reducing bacteria overwhelm the sulfate-reducing bacteria when competing, inducing nitrate in the digester augments sulfide reduction [147]. During the desulfurization process, autotrophic denitrification has been frequently used, in which sulfide oxidizing bacteria employ sulfide as an electron donor to link with the nitrate-lesening process [148]. When sulfide is oxidized to sulfate, elemental sulfur is generated as a transitional product. The process is enhanced by some bacteria species, such as *Thiobacillus denitrificans* which can also reduce nitrogenous species to dinitrogen apart from oxidizing sulfide to elemental sulfur. Below is a summary of the reactions (Eqns. (9)–(12)) that influence the anoxic technique [149]:





Yang and other colleagues [151] presented one of the studies looking at the efficacy of anoxic techniques in reducing sulfide concentrations in wastewater. Various tests were developed to investigate chemical and biological sulfide oxidation through nitrate at the water level. The findings revealed that sulfide oxidation was biologically controlled in anoxic settings, with elemental sulfur being retrieved as a byproduct. However, as an intermediary product, nitrite was deposited in the effluent. Furthermore, Yong Zeng and other co-authors [152], according to their findings, the loading rate can affect sulfide removal efficiency. However, the results show that the removal efficiencies plummeted substantially as the loading rate was reduced. For example, nearly 62% and 50% of efficacies were recounted when biogas slurry was recycled. This exclusion capability is compliant with the outcomes from the experiment, which was done by Mouna and companion researchers to assess the impact of several other parameters, such as the amount of H₂S, Empty Bed Residence Time (EBRT), and the molar ratio of nitrogen to sulfide (N/S), in conjunction with the performance of the biofilter under anoxic conditions.

However, when sufficient NO₃⁻ is present, H₂S is entirely oxidized into sulfate [153,58]. This incident shows how the sulfide to nitrate (S/N) molar ratio is fundamental throughout H₂S oxidation. Referring to the research which was done by Li and other colleagues [154] to investigate the influence of the molar proportion of S/N on biogas desulfurization performance, it was revealed that the expulsion effectiveness of H₂S upgraded from about 66 to 100% upon lessening the S/N ratios from 3.6 to 0.7. In this study, it was then agreed that different approaches for infusing nitrate in wastewater, i.e., intermittent and continuous, did not affect the exclusion of H₂S considerably, while the intermittent inclusion of nitrate wastewater elevated the percentages of sulfate and denitrification performance.

In addition to anoxic bioreactors used for desulfurization, the carbon to nitrogen (C/N) ratio is vital to the AD process. A balanced C/N ratio ensures an appropriate supply of carbon to SRB, which is responsible for converting sulfate to sulfide during desulfurization [155]. Therefore, the C/N ratio not only controls microbial activity but also controls the availability of essential nutrients.

According to Guillermo and other co-authors' study [156], the anoxic desulfurization process improved methane content from 60.0% in the raw biogas to 61.7–63.5% in the purified biogas, increasing the effectiveness of H₂S removal from 92–97% to 92–97%. However, the C/N ratio imbalances may result in process instability due to the accumulation of hazardous intermediates such as volatile fatty acids and an unbalanced pH, which slows the desulfurization process [157,158].

3.3. Biofiltration of hydrogen sulfide (H₂S)

In this process, polluted gas (biogas) is carried into the biofilm and through the biofilter, where microbes use polluting gases such as H₂S and methane (CH₄) as a carbon or energy source. The most common microbial species in H₂S biofiltration is *Thiobacillus* sp., which can break down H₂S for energy and generate sulfate or sulfuric acid [159]. For microbial growth and proper distribution of both air and water, an ideal biofilter media should have a large surface area. As a result, microbial oxidation enables the biofilter to rapidly capture air that is odorless and contains H₂S [102]. Normally, microbial oxidation makes it easier for H₂S in the air to permeate into the biofilm and be broken down by the sulfur oxidizing bacteria (SOB). These microorganisms use H₂S as an energy source during the process to transform H₂S into sulfate (SO₄²⁻) or elemental sulfur [160].

However, biofiltration consistently contributes to decreased volatile fatty acids (VFA) accumulation after H₂S treatment. Depending on the amount and composition of VFA and microorganisms present in the biofilter, the SOB may utilize VFA as a carbon source during the oxidation of H₂S, hence reducing the amount of VFA produced [161,162].

In practice, the efficacy of energy or resource recovery in a treatment system is dependent upon the characteristics of the influent wastewater and process design, such as the capacity of a specific process or material to separate and selectively capture CO₂ and H₂S from a gas mixture, primarily containing CO₂, CH₄ (methane), and H₂S [163]. Since methane can pass through and be recovered as an energy source, activated carbon and zeolites with improved porosity can improve the selectivity of both CO₂ and H₂S [164].

This technique has been considered superior to other conventional methods for evicton of H₂S recently due to its effectiveness in treating lethal pollutants. However, the biofiltration process (as summarized in Table 2) is considered an effective and convenient approach to removing H₂S since it can treat contaminants with very high flow rates regardless of concentration [165].

4. Conclusion and future prospects

4.1. Future prospects of hydrogen sulfide (H₂S) removal

Existing techniques have been modified to enhance biogas production to reduce the impact of H₂S emissions during anaerobic digestion and promote the sustainable utilization of organic waste for energy recovery. Typically, a post-treatment method has been widely employed, which tends to lower the quality of biogas during the process. Nonetheless, this process incurs additional costs, necessitating using absorbent and adsorbent materials to clean the biogas [166]. Among the methods examined in this study, microaeration emerges as a viable and cost-effective alternative, supported by various researchers (Table 3). This method offers advantages over other techniques since it only requires a small amount of oxygen to be introduced into the digester, effectively

Table 3
Advantages and disadvantages of various sulfur control approaches.

Method	Advantages	Disadvantages	Economic considerations based on capital cost ^a	References
Micro-aerobic process	<ul style="list-style-type: none"> - The rate of hydrolysis of both carbohydrates and protein is enhanced by 21–27 and 38–68%, respectively. - Improves the degradation process of COD - No catalyst is required 	<ul style="list-style-type: none"> - If not well controlled can lead to the accumulation of lactic acid. 	Lower operational cost equivalent to 0.0019 EUR/m ³ of biogas treated	[167,168, 55]
Precipitation process	<ul style="list-style-type: none"> - Metals such as Fe and Zn can be recovered for the precipitating process. 	<ul style="list-style-type: none"> - The method requires the use of chemicals, which are hence costly. - Minimum selectivity 	No data	[169,60]
Adsorption process	<ul style="list-style-type: none"> - Adsorbents materials can be functionalized to modify their surface properties for the adsorption process 	<ul style="list-style-type: none"> - The adsorption of H₂S is mostly affected by carbide formation during carbonization. - Limited to the lower concentration of H₂S 	Relatively expensive, with an estimated cost of US\$2.3 per m ³ of biogas	[170,2, 171]
Stripping	<ul style="list-style-type: none"> - It is simple to use 	<ul style="list-style-type: none"> - High energy consumption. - Costs are often involved in building the stripping tower 	No data	[172,173]
Wet scrubbing	<ul style="list-style-type: none"> - Simple to operate - Strong adaptability and high efficiency 	<ul style="list-style-type: none"> - High energy demand for the regeneration process - High corrosiveness of equipment surfaces 	When employing NaOH, it costs roughly US\$2.38/m ³ .	[174,171, 134]
Aerobic biological technology	<ul style="list-style-type: none"> - Energy costs are reduced because the process can take place at ambient temperature 	<ul style="list-style-type: none"> - It is a time-consuming method 	No data	[175,176]
Biofiltration	<ul style="list-style-type: none"> - No harmful byproducts are being generated. - High removal efficiency - Simple to operate. 	<ul style="list-style-type: none"> - The design is not simple and inquires about some operational parameters that need to be optimized. 	High treatment cost nearly US \$1.5/m ³ of biogas	[145,165]

^a Cost data based on estimates for the year 2015–2019 with currency conversion of 1EUR to US \$1.12.

Table 4
The effect of H₂S on equipment and potential solutions.

S/N	Specific damage	Explanation	Potential solution	Reference
1.	Corrosion-Mechanical Attack	H ₂ S can corrode metal surfaces and mechanically harm them, especially when moisture is available. Pipelines, valves, and other pieces of machinery may corrode, resulting in leaks, decreased structural integrity, and, eventually, failure.	<p>F095 Material Selection: H₂S corrosion can be lessened using corrosion-resistant materials like stainless steel or corrosion-resistant alloys.</p> <p>F095F020 Protective Coatings: Protection against corrosive attack can be achieved by coating or lining metal surfaces with the proper materials.</p> <p>F095F020 Cathodic Protection: By applying a protective current to the metal surfaces, cathodic protection devices like sacrificial anodes or impressed current systems may help prevent corrosion.</p>	[41,42]
2.	Failures in Tools and Equipment	H ₂ S exposure can lead to malfunctions in the pumps, compressors, turbines, and electrical components involved in natural gas production. H ₂ S's corrosive properties can damage gaskets, seals, and moving parts, which can cause equipment to break down or malfunction.	<p>F095F020 Regular Inspections and Maintenance: Regular inspections, including non-destructive testing and visual checks, may help detect early indications of degradation or damage and enable prompt repair or replacement.</p> <p>F095F020 Equipment Upgrades: Equipment's resistance against damage from H₂S can be improved by switching to H₂S-resistant components or designs.</p> <p>F095F020 Monitoring and Detection Systems: Installing monitoring systems that can identify H₂S concentrations, provide early notifications, and enable rapid corrective measures can help prevent equipment failures.</p>	[43]
3.	Tool Joint Issues	In drilling operations, tool joints—connections between drill pipes—can also specifically pose problems. H ₂ S can cause embrittlement and stress corrosion cracking in tool joints.	<p>F095F020 Material Selection: The risk of failures can be reduced by selecting tool joint materials with enhanced resistance to H₂S-induced corrosion and cracking</p> <p>F095F020 Enhanced Inspection Techniques: Advanced inspection methods, including ultrasonic or electromagnetic testing, can assist in discovering early indications of destruction in tool joints and enable preventative maintenance or replacement</p>	[44,45]

deactivating sulfate-reducing bacteria that play a key role in the conversion of sulfate ions (SO_4^{2-}) to hydrogen sulfide through assimilation and dissimilation pathways (Fig. 3).

However, for the sustainable and effective implementation of the microaeration method, pretreatment is advisable to achieve satisfactory outcomes in removing H_2S from the digester. In contrast to post-treatment, pretreatment is a relatively straightforward and cost-effective approach. It involves using chemicals to facilitate the precipitation of sulfide (S^{2-}) and sulfate (SO_4^{2-}) ions in the substrate before introducing them into the digester. Recent studies have demonstrated promising results for pretreatment in laboratory-scale applications [177]. However, further research is required to investigate the cost estimation and optimization of chemical addition for desulfurization when applied at a larger, full-scale level.

4.2. Conclusion

This review examines different approaches to controlling H_2S by considering various factors, including operating principles, suitable conditions, economic viability, and the advantages and disadvantages of each technique. Post-treatment methods like bio-filtration and wet scrubbers offer highly effective removal of H_2S , particularly for large-scale applications, although with increased costs. Nevertheless, given the increasing demand for alternative energy sources like biogas, the micro aeration method is considered a straightforward and economical approach to enhance desulfurization while treating sulfur-containing materials for anaerobic digestion. In order to mitigate the risk of excessive aeration, the information presented also highlights the significance of a thorough design framework and the sharing of operational experiences.

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Author contribution statement

All authors listed have significantly contributed to the development and writing of this article.

- 1) conceived and designed the experiments.
- 2) performed the experiments.
- 3) analyzed and interpreted the data.
- 4) contributed reagents, materials, analysis tools or data.
- 5) wrote the paper.

Data availability statement

Data will be made available on request.

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.

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Appendix A. Supplementary data

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