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Sewage sludge management and enhanced energy recovery using anaerobic digestion: an insight

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ABSTRACT

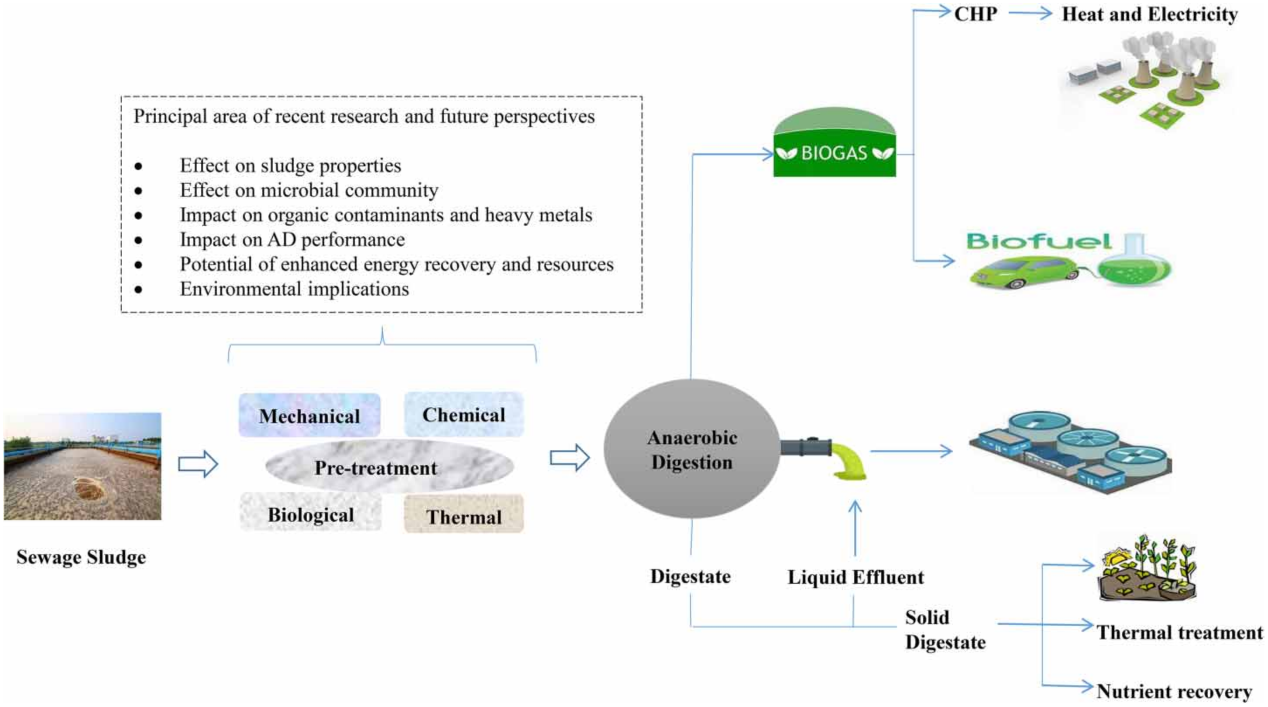
Sewage sludge (SS) is a potential source of bioenergy, yet its management is a global concern. Anaerobic digestion (AD) is applied to effectively valorize SS by reclaiming energy in the form of methane. However, the complex floc structure of SS hinders hydrolysis during AD process, thus resulting in lower process efficiency. To overcome the rate-limiting hydrolysis, various pre-treatment methods have been developed to enhance AD efficiency. This review aims to provide insights into recent advancements in pre-treatment technologies, including mechanical, chemical, thermal, and biological methods. Each technology was critically evaluated and compared, and its relative worth was summarized based on full-scale applicability, along with economic benefits, AD performance improvements, and impact on digested sludge. The paper illuminates the readers about existing research gaps, and the future research needed for successful implementation of these approaches at full scale.

Key words: anaerobic digestion, bioenergy, hydrolysis, sewage sludge, sludge pre-treatment

HIGHLIGHTS

- AD offers sustainable sludge management by harnessing energy recovery.
- Pre-treatment approaches for enhanced hydrolysis during AD.
- Challenges and recent advances during sludge pre-treatments are discussed.
- Further research on detailed characterization after pre-treatment is required.
- The future outlook for pre-treated sewage sludge digestion has been discussed.

GRAPHICAL ABSTRACT



LIST OF NOMENCLATURE

AD	Anaerobic digestion
AS-MBR	Aerobic sludge membrane bioreactor
BMP	Biomethanation potential
CHP	Combined heat and power
COD	Chemical oxygen demand
CSTR	Continuous stirred tank reactor
DD	Disintegration degree
EPS	Extra polymeric substances
EPT	Energy required for pre-treatment
FAO	Food and agricultural organization
GHGs	Greenhouse gases
HPH	High-pressure homogenization
HRT	Hydraulic retention time
MLSS	Mixed liquor suspended solids
OLR	Organic loading rate
OM	Organic matter
PACs	Polycyclic aromatic compounds
PAHs	Polyaromatic hydrocarbons
sCOD	Soluble chemical oxygen demand
SRT	Sludge retention time
SS	Sewage sludge
tCOD	Total chemical oxygen demand
TPAD	Temperature-phased anaerobic digestion
TS	Total solids
TSS	Total suspended solids
VFA	Volatile fatty acid
VS	Volatile solids
VSS	Volatile suspended solids
WAS	Waste-activated sludge
WHO	World health organization

1. INTRODUCTION

Municipal wastewater treatment generates sludge as a byproduct, which is primarily composed of biomass. Generally, the activated sludge process is adopted worldwide for the treatment of municipal wastewater, which yields a massive quantity of sludge. The proper disposal of sludge is challenging due to its large volume, presence of pathogens and foul odor (Świerczek *et al.* 2018). Raw sludge typically contains 2–3% solids, with the remainder being water. In India, approximately 72 million m^3/d of sewage is generated of which 32 million m^3/d is being treated (CPCB 2021). Treating all generated sewage would produce approximately 4,000 tonnes of dry sewage sludge (SS) annually. Dewatered sludge typically comprises organic matter (OM) (50–70%) and mineral components (30–50%), including 1–4% of inorganic carbon. It also contains nitrogen (3.4–4.0%), phosphorus (0.5–2.5%), and significant quantities of various other nutrients (Tyagi & Lo 2013).

Currently, municipalities primarily manage sludge disposal through landfilling, application to agricultural fields, incineration, and anaerobic digestion (AD). However, traditional methods like landfilling, incineration, and agricultural use have faced challenges due to legislative measures and public perceptions (Hassan *et al.* 2023). Worldwide, municipal wastewater treatment plants produce approximately 45 million dry tonnes of SS annually, with significant contributions from the European Union, the USA, and China, which generate between 18 and 33 million tons each year (Gao *et al.* 2020). This volume is expected to rise due to population growth and stricter environmental regulations. In the European Union, sludge disposal methods include incineration (25%), which removes 70% of solids but produces metal-rich ash; landfilling (9%); agricultural reuse (27%), subjected to regulatory permissions; composting (21%); and other methods (18%) (Gao *et al.* 2020; Ragi *et al.* 2022). These disposal options carry considerable costs and have environmental and logistical limitations. In European countries, legislative restrictions have led to bans on the operation and construction of new landfills to prevent contamination from leachates containing heavy metals, posing significant challenges for SS disposal (Wagner & Schlummer 2020). Similarly, applying sludge to agricultural land can be an effective management strategy, but excessive or direct application poses risks. Application of SS in agriculture may lead to contamination of the food chain through the absorption of carcinogens and heavy metals by crops (Qin *et al.* 2021). Additionally, excessive sludge application can result in the bioaccumulation of contaminants, adversely affecting soil microbes (Clarke & Cummins 2015). In areas with unavailability and dense populations, incineration can be used as an alternative method for sludge disposal as it reduces sludge volume. Generally, sludge has a low heating value with high moisture content, thereby requiring high energy consumption in the drying of sludge; thus, the energy intensiveness of incineration facilities poses a significant challenge in its application. Incineration also contributes to the air pollution with greenhouse gases (GHG) and other harmful substances and also creates ash containing hazardous materials (Makarichi *et al.* 2018).

With the surge in energy and fertilizer demand in the global market, organic wastes including SS are viable resource recovery options (Tyagi & Lo 2013). By applying modern and innovative technologies, these substrates can be utilized to generate heat and power. SS has the potential to replace synthetic fertilizer as a soil conditioner after proper treatment using appropriate technologies. In comparison to landfilling, applications of SS for energy and soil conditioners are environmentally sustainable and economical. For sewage treatment, energy consumption is approximately 300–2,100 kWh/million m^3/d (Gandiglio *et al.* 2017). Approximately 50–60% of the total energy in the activated sludge process is utilized in aeration with sludge management requiring 15–25% of total energy (Mamais *et al.* 2015). Using SS as an energy source may also contribute toward minimizing fossil fuel requirements and reducing GHGs (Zhao *et al.* 2023). Economic gain can be achieved by offsetting treatment expenses, reducing health expenditures, and declining energy expenses by consuming biofuels or biogas as a partial replacement for conventional fuels. For effective utilization of bio-solids, proper treatment should be performed. Thus, with the advent of new technologies and advancements in existing ones, there are opportunities for greater energy recovery and the reduction of risks associated with pollutants in wastewater.

Currently, wastewater treatment plants (WWTPs) are integrated with anaerobic sludge digestion units. This technology is economical and efficient as it provides energy recovery in the form of biogas, and the digested sludge can be used in agricultural fields as a source of nutrients, thus reducing environmental impact (Caiardi *et al.* 2022). However, due to certain factors, such as cell lysis, extra polymeric substances (EPS), and complex compounds in sludge, the efficiency of the process is compromised (Khanh Nguyen *et al.* 2021). To overcome these issues associated with the AD process, the integration of pretreatment methods becomes a necessity.

Recently, a significant emphasis has been on boosting energy and resource recovery from sludge. Various pre-treatments, including mechanical and chemical methods, can be implemented to enhance the efficiency of the AD process (Atelge *et al.*

2020). These pre-treatments can also facilitate resource recovery from sludge. To conduct a thorough examination of the literature on SS and energy recovery, a comprehensive literature review was carried out using bibliometric analysis (Supplementary information). The novelty of this review paper lies in its comprehensive examination of SS management practices, with a particular emphasis on energy recovery. This paper delves into the intricate aspects of sludge utilization and offers a holistic approach to sludge management. Furthermore, the paper thoroughly explores various thermal, physical, and chemical pre-treatment methods aimed at enhancing biogas generation through AD. A notable contribution of this paper is the inclusion of a statistical analysis, which compares the effectiveness of all three pre-treatment techniques (thermal, physical, and chemical) in terms of solubilization increase and methane increment. This rigorous literature analysis allows for the determination of which pre-treatment method is most effective in augmenting biogas generation, providing valuable insights for future research and practical applications in SS management.

2. SS MANAGEMENT OPTIONS

A diverse range of technologies and treatments are available for the safe disposal of sludge post-resource and energy recovery. The most common methods that are adopted for SS include landfilling and direct application to agricultural fields, which are less energy-intensive but have negative environmental and human health impacts. Energy and resources from SS can be obtained using various technologies, including AD and thermal treatments, such as incineration, pyrolysis, and gasification. These technologies aid in holistic SS management with less adverse environmental impacts to a more significant extent. The current SS management practices are further discussed in detail in upcoming sections.

2.1. Application in agriculture and soil reclamation

SS can be utilized in agricultural fields after ensuring the chemical and biological properties of sludge. Approximately 40% of the total sludge is applied on land or used in agriculture (Roig *et al.* 2012). Contaminants like polycyclic aromatic compounds (PACs), phenols, grease, and heavy metals in SS can create severe alterations in soil by disturbing flora and fauna and reducing soil fertility. The excessive and uncontrolled application of sludge may cause detrimental effects on soil and groundwater by seepage of contaminants (Houillon & Jolliet 2005). The characteristics of soil, such as pH, cation exchange capacity, OM composition, and heavy metals, alter with the addition of SS to the soil (Singh *et al.* 2020). During sewage treatment, some part of heavy metals may be sequestrated in the organic part of sludge due to the formation of complexes (Florentino *et al.* 2019).

Zhang *et al.* (2022) assessed the impact of sludge application in agriculture using lettuce as an experimental crop. At a dose of 0.2 g/kg, crop yield increased while heavy metals remained within safe limits. However, increasing the dosage to 8 g/kg resulted in the contamination of crops and soil with heavy metals. The application of SS in forestry, particularly on infertile tropical soils, has significantly enhanced the growth of Eucalyptus trees. Notably, a single application of SS demonstrated a residual effect lasting even after 10 years, raising concerns about the potential accumulation of toxic contaminants in the soil (Florentino *et al.* 2019). Dhanker *et al.* (2021) studied the effect of different doses of SS amendment on soil and found that SS application up to 50 tonnes/hectare improves soil nutrient and organic content. However, higher doses increase the enzymatic activities in soil along with an increase in heavy metals. The contamination due to heavy metal accumulation is a threat in almost all land applications of SS (Cieślik *et al.* 2015). It is vital to control the concentration of contamination in soils when using processed sludge for agricultural and reclamation purposes. The sludge should be treated before reuse, depending upon the application in forestry, agriculture, or land reclamation. Adequate treatment by disinfecting and stabilizing sludge by earthworms or drying beds, either in combination with modification of other technologies, becomes a requisite for land application of sludge (Suthar 2010). While implementing SS for agricultural or land applications, the dose of sludge and the characteristics of sludge have to be ensured to avoid negative impacts on the soil. In the past three decades, concerns about the safe reuse of treated wastewater and SS in agriculture have led to the establishment of international and local regulations. These guidelines and regulations, set by international organizations and local environmental agencies, aim to ensure safe practices (Catenacci *et al.* 2022a). Typically, these standards take one of three approaches: limiting the concentration of pollutants allowed in the sludge, setting a maximum permissible level of pollutants in the soil after application, or restricting the total amount of sludge or pollutants that can be applied to the land (Mabrouk *et al.* 2023). Food and Agriculture Organization (FAO) guidelines and the European Directive 86/278/EEC set limits for toxic elements in soil after applying SS and limit the quantity that can be added each year (Nunes *et al.* 2021). A report by Chang *et al.* (1995) for the World Health Organisation (WHO) outlines the maximum levels of pollutants (organic and inorganic) permissible in soil when using treated wastewater or SS for irrigation. However, the report also acknowledges that specific limits may vary depending on local regulations.

2.2. Energy recovery

There are different ways of utilizing SS. The direct application of sludge in soil and agriculture may have negative impacts, such as the accumulation of heavy metals in the soil and crop. Energy recovery from sludge can also be explored as an option for sludge management. Various methods exist for energy recovery from sludge treatment, including heat and bioenergy. Commonly applied methods fall into two categories: biological and thermo-chemical. These encompass techniques, such as AD, incineration, combustion, pyrolysis, and gasification. AD is a biological route for sludge treatment, producing bioenergy in the form of methane (Wang *et al.* 2021). The application of microbes for biodegradation is a multistage, time-consuming process. The biogas generated from this process comprises 60–70% methane, 30–40% carbon dioxide, and trace elements of other gases (H_2S) (Zhen *et al.* 2017). The process is simple and effective, with ease of operation and maintenance compared to thermal treatment, which has the drawback of being time-consuming.

Thermo-chemical treatments generally have shorter reaction times but require sludge with lower moisture content. The process comprises sludge combustion in the presence or absence of oxygen to produce heat. This technology is well-established and offers heat and electricity generation potential. The other benefits of thermo-chemical techniques, such as pyrolysis and gasification, include producing bio-oil and syngas, which can be used as fuel (Hu *et al.* 2022). The drawback of this technology associated with SS treatment is the high energy requirement due to higher moisture content, GHG emissions, and ash disposal. Due to their operation at high temperatures, these processes entail significant initial investment as well as operational and maintenance expenses. Improper operation can result in the formation of harmful by-products (Ding *et al.* 2021).

2.2.1. Thermal processes

To treat sludge using thermal technologies, such as pyrolysis, incineration, and co-incineration, pre-treatment of sludge is required to reduce moisture content. Drying sludge with the help of heat-generating microbes (biodrying) can be an economical alternative (Gao *et al.* 2020). Methane fermentation can also be performed to recover energy before drying sludge. The final product achieved after drying can be further reused depending on the calorific value. The low calorific value end product can be utilized in the construction industry, such as road ballast and pellets, whereas high calorific value end product can be used for fuel and energy recovery purposes (Hu *et al.* 2022).

Incineration is one of the commonly used thermal treatment processes applied for managing different types of wastes, such as municipal and medical wastes. Incineration offers significant volume reduction for SS. This characteristic makes it a viable disposal option in densely populated regions with limited land availability and high sludge generation rates (Samolada & Zabaniotou 2014; Liang *et al.* 2021). Before incineration, SS must be dried with 18–35% dry solids, generally about 25% solid content is required for sludge to burn auto-thermically (Houillon & Jolliet 2005; Donatello & Cheeseman 2013). Proper design makes these systems economical and has fewer operational costs (Winkler *et al.* 2013). The high temperatures generated during SS incineration present an opportunity for cogeneration. This captured heat could be used for heating in nearby buildings or for on-site pre-drying of sludge before incineration, further increasing process efficiency. Meanwhile, there is also a good possibility of using this heat for the production of clinkers (Valderrama *et al.* 2013; Schnell *et al.* 2020). Generally, the waste product achieved after incineration is ash, which can be reused or utilized for further use. Ash generated from various gas filters, including cyclone filters, bag filters, wet scrubbers, etc., may vary in composition. Consequently, the management method for the ash may differ and could be interconnected (Hu *et al.* 2021).

Pyrolysis converts waste-activated sludge (WAS) to solid residue (char) while producing bio-oil in the absence of oxygen. The quality and quantity of by-products depend upon SS operating temperature characteristics, time, and pressure (Tian *et al.* 2013). Compared to incineration, pyrolysis reactions require energy of around 100 kJ/kg (Khiari *et al.* 2004). In a study by Gao *et al.* (2017), bio-oil obtained from the pyrolysis of WAS showed low heavy metal concentrations, as most of the metals were retained in the char. Although pyrolysis produces a substantial amount of char with potential uses as fuel, adsorbent, and soil conditioner, the issue of heavy metals contamination remains. The limiting factors in pyrolysis implementation for WAS management are the economic feasibility and requirement of complex equipment (Manara & Zabaniotou 2012; Shahbeig & Nosrati 2020). Another useful method to process a significant amount of organic waste involves combining AD with pyrolysis. This integrated approach efficiently converts organic material from waste to valuable products (Pecchi & Baratieri 2019; Caiardi *et al.* 2022). The combined approach effectively resolves the issue of AD digestate disposal by converting it into biochar, thus enhancing waste management sustainability (González-Arias *et al.* 2020). AD coupled with pyrolysis displays higher apparent energy efficiency (71.4%) compared to pyrolysis (60.4%) (Cao & Pawłowski 2012). Similarly, Monlau *et al.* (2015) achieved a net energy increase of 42% for AD-coupled pyrolysis. However, Li & Feng (2018) reported that AD outperformed both pyrolysis and combined systems, except for sludge with high organic content. The development of these integrated systems is relatively recent and highlights the need for further research to assess their technical, economic, and environmental sustainability (Tayibi *et al.* 2021).

Gasification is another technology that converts WAS to combustible gases (syngas) and keeps the heavy metals and other contaminants fixed in solid residue (Roche *et al.* 2014; Sikarwar *et al.* 2016). However, gasification also faces significant challenges due to lower heating values and the high moisture content of WAS. Another major obstacle in the gasification of WAS is large tar production, which requires further treatment, thus making the process economically and technically non-appealing (Kokalj *et al.* 2017).

Co-incineration becomes a requisite for SS due to its low calorific value. For co-incineration, low calorific sludge is mixed with fuels, such as coal or gases (Donatello & Cheeseman 2013; Schnell *et al.* 2020). While using SS as an energy source, the calorific value of SS should be considered in addition to economics, as the final product may end up with contaminants. Synergy with industries can be beneficial if incinerators or higher calorific value products are produced near treatment plants (Liu *et al.* 2016). Co-incinerating municipal solid waste and SS can be environmentally and economically safe (Lin & Ma 2012). However, the ash produced from each incineration facility must be examined for chemical toxicity and ecotoxicity (Barbosa *et al.* 2011; Hong *et al.* 2013; Magdziarz & Werle 2014). Generally, coal ash contains fewer nutrients than SS ash; thus, different management methods should be adopted for ash generated from different sources (Donatello & Cheeseman 2013). To prevent pollution, emissions should meet the discharge criteria and be free from pollutants, such as polyaromatic hydrocarbons (PAHs) adsorbed on the surface of dust (Liang *et al.* 2021). Ash usually consists of a substantial volume of biologically available phosphorus that can be used on land and in agriculture. For instance, if sludge comes from treatment plants treating industrial wastewater, then the sludge may contain large concentrations of heavy metals that can deteriorate the environment (Gao *et al.* 2020). It is also noteworthy that excessive application of sludge ash may increase the phosphate concentration excessively, thereby leading to the problem of eutrophication in the nearby waterbodies. There is also a possibility of an increase in toxicity due to the application of ash, as PAHs may be adsorbed on the surface of ash particles (Hušek *et al.* 2022). If the products of the sludge managing process are to be applied in agriculture, regulation is required on both the application and fate of these products in the environment.

2.2.2. Anaerobic digestion

AD is one of the oldest biological processes that convert complex OM to simple form in the absence of oxygen (Lohmeyer 1959; Agabo-García *et al.* 2019; Pan *et al.* 2019). During the AD process, diverse microbial communities transform the OM to give resource-rich material and biogas (Lastella *et al.* 2002). The key objective of AD of SS is the reduction of pathogens along with energy recovery, and this is achieved by providing anaerobic conditions for converting OM to methane and carbon dioxide (Zhao & Liu 2019). Various microbial species perform different enzymatic responses for methane production by degradation of OM (Chen *et al.* 2020b). AD is a complex process that has four stages: hydrolysis, acidogenesis, acetogenesis, and methanogenesis. Complex compounds such as lipids, proteins, polysaccharides, and other OM are transformed into amino and fatty acids in the hydrolysis step of AD. However, this step is reflected as a rate-limiting step of the AD process (Appels *et al.* 2008a). The hydrolysis and acid fermentation microbes include obligate anaerobes and facultative microorganisms (Zamorano-López *et al.* 2020). Various genera recognized in anaerobic digesters include *Clostridium, Corynebacterium, Actinomyces, Staphylococcus*, and *Escherichia*. The conversion of methane is performed by *Methanosarcina, Methanothrix, Methanococcus, Methanobacterium*, and *Methanoculleus* microbes (Liew *et al.* 2022). For the production of methane and carbon dioxide, acetate is utilized by *Methanosarcina* and *Methanothrix*, whereas oxidation of hydrogen with carbon dioxide acting as an electron acceptor is performed by *Methanococcus, Methanobacterium*, and *Methanoculleus* (Senés-Guerrero *et al.* 2019; Walter *et al.* 2019).

AD is an environment-friendly technology, although this technology faces the challenge of a high retention period and lower removal efficiency for organic compounds (Park *et al.* 2005). Additionally, different parameters, such as carbon/nitrogen ratio, nitrogen, temperature, alkalinity, and pH affect the process efficiency (Andreoli *et al.* 2015). Sludge consists of complex and high amounts of OM in particulate form, which takes time to degrade. Pre-treatment of sludge is recommended to reduce the particle size and rupture biomass's cell wall, thus accelerating the solubilization of particulate matter (Amani *et al.* 2010). With pre-treatment, the cell lysis occurs at a faster rate, digestion time is reduced, volatile solids (VS) degradation is quick, and the amount of digestate is reduced after AD compared to conventional AD (Neumann *et al.* 2016). Generally, AD without pre-treatment causes a lower VS content reduction (30–50%) with a longer retention time of 20–30 days at mesophilic conditions (Appels *et al.* 2008a). Therefore, pre-treatment technologies have been developed to minimize the reaction time and completely utilize the OM in digested sludge (Atelge *et al.* 2020). The application of pre-treatment methods before AD can lead to improvements in biogas quality, reduction in retention time, and enhancement of overall process efficiency.

3. PRE-TREATMENT OPTIONS TO REDUCE SLUDGE VOLUME AND ENHANCE METHANE RECOVERY

Pre-treatment comprises the combination of different physico-chemical processes to enhance the efficiency of AD. The microbial cells are made up of EPS and cell walls, which are difficult to disrupt and limit the hydrolysis rate (Walter *et al.* 2019). Therefore, pre-treatment disrupts cell walls and EPS, thus releasing nutrients and OM to enhance the activity of microbes and improve methane generation. The main aim of the application of pre-treatment includes enhancing sludge biodegradability, improved hydrolysis rate, high methane yield, enhanced dewaterability, and reduced sludge viscosity. This section briefly reviews various pre-treatment approaches, including mechanical, thermal, chemical, biological, and several combinations of processes to enhance AD efficiency.

3.1. Mechanical pre-treatment

In mechanical treatment, the size of particles is reduced to enhance the surface area, thereby aiding in the AD process (Atelge *et al.* 2020). Different researchers have established reduced biogas production due to lower chemical oxygen demand (COD) values associated with large particles (Zamanzadeh *et al.* 2017; Nabi *et al.* 2020; Shabbirahmed *et al.* 2023). Therefore, mechanical pre-treatment can be utilized to reduce the sludge particle size, enhance the OM, and improve the sludge digestibility, as presented in Table S1. The following sections further discuss the most commonly applied mechanical pre-treatments in detail.

3.1.1. Ultrasonic pre-treatment

Ultrasonication is the most effective and well-renowned technology for improving sludge biodegradability. In this method, the sludge structure is disrupted by developing hydro-mechanical shear forces due to the formation of microbubbles. The collapsing of these microbubbles induces cavitation and disrupts sludge cell structures (Kavitha *et al.* 2019; Atelge *et al.* 2020). During sludge pre-treatment, cavitation is influenced by various physical factors like density, temperature, and ultrasound frequency (Pilli *et al.* 2011; Li *et al.* 2018).

Ultrasonication alters the sludge biologically, chemically, and physically by simulating biological activity, the release of enzymes, solubilization of organic compounds, and particle size reduction (Pilli *et al.* 2011; Guo *et al.* 2013). Ultrasonication can also alter the COD particle size distribution, shifting the peak from the particulate fraction ($>$1,600 nm) to the smallest size range ($<$2 nm) (Doğruel & Özgen 2017). The time and frequency of sonication vastly affect the efficiency of AD. The cavitation effect, defined by the critical size of cavitation bubbles, is influenced by the ultrasonic frequency, typically between 20 and 25 kHz. While sludge solubilization increases with prolonged ultrasonic treatment, it should not exceed 15 min, as the improvement in solubilization diminishes beyond this duration (Bhat & Gogate 2021). For example, the SCOD increase was approximately 45.6% at 15 min, but it reached 67.9% at 240 min (Şenol 2021). Ultrasonic pre-treatment can liberate recalcitrant organics like humic substances and high-molecular-weight proteins into the liquid phase. These compounds may inhibit coenzyme F420, a crucial enzyme in methanogenesis (Zheng *et al.* 2023). Additionally, the concentration of these inhibitory substances increases with longer treatment times (Lu *et al.* 2018). Sonification performed for 80 min at 20 kHz frequency and 0.5 W/mL density by Li *et al.* (2018) displayed WAS dewaterability deterioration and a swift decline in *Methanocorpusculum* abundance with a 53.8% enhancement in methane generation. Appels *et al.* (2008b) also showcased the effect of ultrasonication at different levels of energy input and displayed a 40% increment in biogas production at low specific energy feed compared to a 15% increment at moderate specific energy input. Pre-treatment of WAS with ultrasonication also provides the benefits of reduction in WAS quantity, improved dewaterability, and enhanced release of COD from sludge (Shabbirahmed *et al.* 2023). Based on the literature review, ultrasonification is the most applied method for improving dewaterability and enhancing biogas generation during AD of WAS (Oz & Yarimtepe 2014; Le *et al.* 2015; Li *et al.* 2018). However, due to energy requirements, the main challenge associated with ultrasonication is high cost, limiting the application of this promising technology.

3.1.2. High-pressure homogenization

High-pressure homogenization (HPH) is a process that involves sudden pressure rise up to 900 bars, leading to the development of high turbulence, large shear force and cavitation, and subsequent depressurization. The abrupt changes in pressure during HPH result in the hydrolysis of macromolecules and the rise in soluble COD (sCOD) (Salihu & Alam 2016; Nabi *et al.* 2019). Zhang *et al.* (2012) perceived that for SS with 2.8% total solids (TS), single cycle and homogenization pressure of 30 MPa was the most energy efficate treatment. Whereas, for sludge with 4.2% TS, with four cycles for homogenization and pressure at 80 MPa, a maximum sludge deterioration of 43.94% was achieved (Zhang *et al.* 2012). Nabi *et al.* (2019) investigated the influence of pressure increment from 20 to 60 MPa, displaying sSCOD increment from 1,053 to 2,342 mg/L, methane yield from 77 to 150 mL/g VS, and methane content increment from 47 to 64%. The solid content of SS is recommended below 25 g/L TS for optimal results (Nabi *et al.* 2020, 2021). The key advantages of HPH are odor reduction in reactor headspace during municipal sludge digestion and enhanced biogas production (Wahidunnabi & Eskicioglu 2014). However, HPH does not significantly affect pathogen elimination during AD (Khanh Nguyen *et al.* 2021).

3.1.3. Microwave irradiation

Microwave irradiation (MI) has also been applied as a pre-treatment method prior to AD of WAS. Operation of the pre-treatment occurs at 1 mm to 1 m wavelength and frequency of 300 GHz and 300 MHz (Aguilar-Reynosa *et al.* 2017). It has been reported that with OM solubilization, biogas generation is enhanced by up to 50% for microwave pre-treatment (Beszédes *et al.* 2011). Moreover, in the semi-continuous mode, methane yield was enhanced by 20% with a 70% increment in biodegradability of WAS post-microwave treatment (Gil *et al.* 2018). Primary and secondary sludge mixtures subjected to MI before AD reduced volatile solids (VS) reduction by 41% and 3.2 times augmentation in sCOD (Park & Ahn 2011). The key benefit of MI and enhanced biogas generation is the destruction of pathogens in AD. Irradiation at 70 °C (900 W; HRT = 15–25 days) prior to AD reduced the amount of *Clostridium perfringens* by 50%, total bacteria by 77%, and *Salmonella* spp. and *Escherichia coli* by 100%. In addition, methane production for pre-treated sludge compared to raw sludge was improved by 35% (Kuglarz *et al.* 2013). Moreover, the thermal effect plays a more significant role in OM dissolution in microwave treatment than the pre-treatment duration (Kor-Bicakci *et al.* 2019). Despite an increase in microwave temperature, a corresponding rise in methane yield during AD might not always be observed (Bozkurt & Apul 2020). This could be attributed to the formation of more recalcitrant organic compounds due to the thermal effects of the treatment (Kor-Bicakci *et al.* 2019).

3.1.4. Electro-kinetic disintegration

The electro-kinetic method applies high voltage for the disruption of biomass cells and their structure. It is also known as an electric pulse due to the application of a pulsing electric field (20–30 kV) (Zhen *et al.* 2017). The effect of 34 kWh/m^3 electric pulse treatment on AD of WAS, evidenced a 110–460% improvement in soluble compounds with an 18% surge in the removal of total COD (tCOD) and 10–33% enhancement in methane generation (Lee & Rittmann 2011). Similarly, for pulse treatment at 19 kV and 110 Hz for 1.5 s, the sCOD/tCOD ratio improved by 4.5 with 2.5 times improvement in biogas generation by AD (Choi *et al.* 2006). Electro-kinetic disintegration prior to AD also significantly alters bacterial diversity. Zhang *et al.* (2009) applied an electro-kinetic method on sludge and observed microbial species alteration and methane production improvement by 30%. The conditions and equipment for electric decomposition vary widely across studies (Wang *et al.* 2023), making it difficult to standardize process parameters like voltage, current density, and electrode distance. Further research is needed to ensure pre-treatment effectiveness and operational safety for engineering applications. The critical challenges associated with this technology are energy-intensive as the energy required during pre-treatment is not offset by the increment in methane production, thus making the technology non-viable on a large-scale.

3.2. Chemical pre-treatments

Pre-treatment using chemicals is the most promising method for deleting complex OM. It involves the utilization of chemical processes such as acid and alkaline hydrolysis, ozonation, and other oxidation methods (Wang *et al.* 2020). The pre-treatment enhances the biodegradability of cellulose, thus improving biogas production (Yang & Wang 2019). The efficacy of chemical pre-treatment depends upon the method applied, the chemicals used, and the OM characteristics of the feed. The upcoming section discusses the various chemical pre-treatments and their effects on the improvement of AD, as presented in Table S2.

3.2.1. Alkali pre-treatment

It is one of the most common methods applied for disintegrating biomass and EPS in sludge. After alkali treatment, the acetate structure of the feedstock is eliminated, thus making the material readily available for hydrolysis (Karp *et al.* 2015). The hydroxyl radicals released cause the destruction of cells along with an increment in surface area due to salvation and saponification, thereby increasing the availability of substrate (Carlsson *et al.* 2012). With the increase in pH, the proteins and lipid saponification in the substrate lose their form. As the cell wall cannot sustain turgor pressure, the EPS is destroyed, and intracellular material is released (Banu & Kavitha 2017). Thus, the solubilization efficiency of this method is really high; however, it is affected by the type of alkali used (Behera *et al.* 2014). Generally, NaOH, KOH, $Mg(OH)_{2,}$ and $Ca(OH)_2$ are used for solubilization, with monobasic alkalis more effective than dibasic alkalis (Maryam *et al.* 2021; Toutian *et al.* 2021). The dosage of chemicals governs the solubilization efficiency. Higher dosages achieve higher solubilization but hamper the AD process (Lin *et al.* 2009; Fang *et al.* 2014). The addition of alkali to the SS improves biogas production during AD as it enhances the concentration of volatile suspended solids (VSS) after alkali treatment (Hu *et al.* 2009; Wei *et al.* 2010; Li *et al.* 2012; Lorenci Woiciechowski *et al.* 2020).

Alkali treatment also contributes to an increase in soluble macromolecules, such as proteins and carbohydrates along with COD solubilization, contributing to the higher yield of biogas during AD (Xu *et al.* 2014). After alkali pre-treatment, Xu *et al.* (2020) found 9.82 times increments in soluble protein, 12.16 times in polysaccharide, and 16.11 in sCOD at a pH of 12 along with a 50% increment in methane compared to the control. The application of alkali pre-treatment enhances sludge's dewaterability and helps dispose of WAS (Shao *et al.* 2012). Additionally, post-alkaline treatment pathogens like *Escherichia coli*, viable helminth eggs, and *Salmonella* spp. were eliminated, whereas *Azospira oryzae*, *Dechloromonas denitrificans, Geothrix* spp., and *Geobacter* spp. persists even at $pH > 12.0$ (Lopes *et al.* 2020). The end products of alkaline pre-treatment are non-toxic and contain highly solubilized OM, with the process being less energy-intensive than that of normal conditions (Lopes *et al.* 2020). However, the main challenge associated with alkaline treatment is residual chemicals that inhibit the AD process and the high cost of alkaline and alteration of lignin structure (Brodeur *et al.* 2011; Kim *et al.* 2013).

3.2.2. Acid pre-treatment

This method is more suitable for the treatment of sludge containing lignocellulosic matter as it supports the breakdown of lignin and hydrolytic accumulation of microbes under an acidic environment (Mussoline *et al.* 2013). The main factor in acid treatment is pH, which governs the solubilization of tCOD and other macromolecules present in sludge and enhances biogas production during AD (Devlin *et al.* 2011; Malhotra & Garg 2019). Acid pre-treatment also aids in enhancing hydrogen-producing bacteria along with the boost in methane generation (Tommasi *et al.* 2008). However, inhibiting by-products, such as hydroxymethylfurfural and furfural may be produced by the application of concentrated acids (Jönsson & Martín 2016). Using concentrated acid for acid pre-treatment is not preferred due to its corrosive nature, and it also escalates cost because of the need for neutralization before further treatment (Bhatt & Shilpa 2014; Lee *et al.* 2019). The drawbacks of acid pre-treatment include the formation of inhibitory products, such as hydroxymethylfurfural, the potential loss of simple sugars due to increased degradation, and the necessity of pH neutralization before AD (Wang *et al.* 2020).

3.2.3. Ozonation

Ozonation has received wide attention in the pre-treatment of WAS due to ozone (O_3) being a strong oxidizing agent. This method has the advantages of not leaving any chemical residue and incrementing salt concentrations after pre-treatment (Hartmann *et al.* 2010). Ozone has two mechanisms (direct and indirect) of reactions with OM. Indirect ozone reaction occurs based on hydroxyl radicals, whereas rapid ozone decomposition into radicals causes the direct ozone reaction with sludge. The direct reaction mechanism is an oxidation reaction governed by the substrate combination. These mechanisms diffuse OM into the liquid by disseminating fine particles of the substrate (Yukesh Kannah *et al.* 2017). The efficiency of this process is governed by reactant structure, which aids in making recalcitrant matter more biodegradable (Waring & Wells 2015). Ozone pre-treatment effectively enhances the hydrolysis of organic components in SS, including proteins, polysaccharides, and lipids (Khanh Nguyen *et al.* 2021). A principal component analysis suggested that ozone treatment primarily targets aromatic proteins and their constituent amino acids, such as tryptophan and tyrosine. However, it also increases fulvic and humic acids, especially with longer treatment times and higher ozone doses (Du *et al.* 2021). Ozonation prior to AD can improve biogas yield by 200%, even at mild ozone treatment (Bougrier *et al.* 2007; Ak *et al.* 2013). In a study by Catenacci *et al.* (2022b), the methane yield (128–204 mL/g VS) increased with increasing ozone dosage (0–90 mg O_3/g VS). However, further increases in ozone dosage beyond this range did not significantly increase methane yield (Catenacci *et al.* 2022b). Studies have shown that the ideal ozone dosage for sludge solubilization can range between 0.05 and 0.5 g O_3/g TS depending upon the initial properties of the sludge and the specific pre-treatment conditions used (Salihu & Alam 2016). Tuncay *et al.* (2022) reported the highest average daily removal efficiencies for COD (35%), TS (32%), VS (42%), TSS (60%), and VSS (69%), with a 48% increase in methane production using ozonation at ozone dose of 0.06 g O_3/g TSS. Ozonation is also effective for pathogen elimination and enhanced solubilization of sludge and biogas production (Wang *et al.* 2018). The major drawback associated with the ozonation process is the instability of ozone and high energy requirement, thus making this process unsuitable for large-scale applications.

3.3. Thermal pre-treatment

The utilization of high-temperature conditions to enhance the digestibility and hydrolysis of SS and other wastes containing OM is considered thermal treatment, as presented in Table S3. The operating conditions for thermal pre-treatment include temperatures up to 220 °C at a pressure of 2–9 bar for the duration of 15–1,200 min (Bochmann & Montgomery 2013). Generally, depending upon operating conditions, thermal pre-treatment may be classified as low-temperature or high-temperature thermal pre-treatment. Thermal pre-treatment applied at a temperature below 100 °C is termed as low-temperature pre-treatment (Chen *et al.* 2020a). This method improves the biodegradability of sludge by stimulating thermophilic microbes and OM solubilization. Temperature, pressure, and treatment time are three major governing parameters for the thermal process. The heat applied for treatment results in the alteration of substrate structure, leading to increased waste biodegradability (Zhou *et al.* 2015). The cellular bonds are destroyed by thermal pre-treatments, thus releasing the cell material into the liquid. During thermal treatment, the substrate swells after the effect of pressure, temperature, and water, causing enhancement in surface area for contact of substrate and microbes. This increment in surface area permits shorter HRT of AD and a decrease in digester size (Xiao *et al.* 2020). Thermal pre-treatment is effective in OM solubilization and for pathogen elimination, odor removal, improved dewaterability and volume reduction of sludge.

Thermal treatment up to 70 °C is effective for improvement in the solubilization of the solids; however, the destruction of pathogens requires a higher temperature (De los Cobos-Vasconcelos *et al.* 2015; Liao *et al.* 2016; Nazari *et al.* 2017). Nazari *et al.* (2017) revealed that 80 °C, 5 h, and pH 10 were optimal for the treatment as sCOD enhanced to 18.3 ± 7.5% and VS declined to 27.7 ± 12.3%. These results specified that OM solubilization is favored by alkaline pH, long reaction time, and higher temperatures. The production of biogas was improved with the increase in temperature during the thermal treatment of SS (Liao *et al.* 2016; Neumann *et al.* 2016; Perendeci *et al.* 2020).

In high-temperature thermal treatment, the temperature applied for the treatment of sludge is more than 100 °C. At higher temperatures, physical disintegration is typically promoted for OM solubilization (Iglesias-Iglesias *et al.* 2019; Mahdy *et al.* 2020). WAS becomes slowly and readily biodegradable after the thermal treatment between 125 and 175 °C (Jo *et al.* 2018). In a mixture of WAS and primary sludge, the optimal temperature range for OM solubilization was 175–200 °C. During treatment periods of 60–120 min and 60–240 min, the COD solubilization ratio improved from 11.25 to 15.1 and 25.1%, respectively (Zhen *et al.* 2017). Thermal pre-treatment was applied from 60 to 210 °C. The results showcased improvement in sludge solubility and methane yield up to 190 °C. Thermal treatment beyond 190 °C revealed to have a negative impact on sludge biodegradability (Climent *et al.* 2007). Zhang *et al.* (2018) displayed the effect of thermal pre-treatment on sludge dewaterability along with an increment in biogas production of high solid digested sludge. Although thermal treatment boosts biogas yield, drawbacks, such as the formation of inhibitors through Maillard reactions and melanoidin formation, coupled with high heating demand due to elevated temperature requirements, constrain the suitability of thermal pre-treatments (Gahlot *et al.* 2022).

3.4. Biological methods

Biological pre-treatments are economical and environment-friendly techniques that utilize microbial communities to enhance the hydrolysis stage of AD. Biological pre-treatments are time-consuming and require optimum conditions to escalate microbes (Meegoda *et al.* 2018). Biological pre-treatment generally consists of micro-aeration, temperature-phased AD, and enzyme-assisted pre-treatments.

3.4.1. Micro-aeration pre-treatment

Micro-aeration treatment assists in the hydrolysis of complex OM by injecting oxygen into the system, which enhances the actions of endogenous microbial communities during hydrolysis. Under anaerobic conditions, certain recalcitrant compounds may remain undegraded, but with micro-aeration, exoenzymes are excreted, thereby stimulating the degradation of these recalcitrant compounds (Lim & Wang 2013). Furthermore, combining oxygen with high temperatures (<70 °C) stimulates the production of hydrolytic enzymes (e.g., proteases) by hydrolytic microbes. During AD, the degradation of organic compounds is facilitated by hydrolytic enzymes, which enhance sludge solubilization. This treatment method is sometimes referred to as the autohydrolytic method (Brémond *et al.* 2018). Various works have showcased the competency of micro-aeration pre-treatment for increased AD hydrolysis and improved methane generation (Montalvo *et al.* 2016). Short-term oxygen treatment positively affects methane yield (Ahn *et al.* 2014; Rashvanlou *et al.* 2021). Aerobic thermophilic bacteria used in AD for treatment of mixed sludge at 55 °C reported a 12% increment in biogas yield and a 27–64% reduction in VS (Jang *et al.* 2014). For micro-aeration treatment of SS, optimization of time, temperature, along with aeration rate was performed by Montalvo *et al.* (2016) with optimum conditions of 48 h, 35 °C, and 0.3 volume per minute (vvm). It was reported that methane yield improved by 111% under optimal pre-treatment conditions compared to conditions without pre-treatment. In another research, a thermophilic proteolytic bacterium *Bacillus licheniformis* was the most effective for OM stabilization and gas generation (Merrylin *et al.* 2013). Overall, it can be established that the micro-aeration treatment applied to SS improves the hydrolysis process in AD and enhances biogas generation.

3.4.2. Temperature-phased anaerobic digestion

In temperature-phased anaerobic digestion (TPAD), sludge hydrolysis prior to AD is performed in two stages at different temperatures. Hydrolysis and acetogenesis are improved in thermophilic conditions, whereas acetogenesis and methanogenesis activities are enhanced in mesophilic conditions (Zhen *et al.* 2017; Brémond *et al.* 2018). Along with the improvement in AD, TPAD has the advantages of being economical, less energy-intensive, and eliminating pathogens (Riau *et al.* 2010). The increase in temperature during AD enhanced the methane yield (Bolzonella *et al.* 2007; Akgul *et al.* 2016; Hameed *et al.* 2019), with improvement in methane generation directly proportional to the temperature. The optimum conditions for improved biodegradability and enhanced methane yield after TPAD were a pH of 6–7 with HRT of 1–2 days at 65 °C by Ge *et al.* (2011). Therefore, with these studies, it can be established that TPAD is an effective pre-treatment for improvement in hydrolysis of sludge along with VS reduction and improvement in biogas yield.

3.4.3. Enzyme-assisted pre-treatments

Recently, enzyme-assisted pre-treatment has gained significant attention in the pre-treatment of sludge prior to AD. This pre-treatment aids in sludge solubilization along with the degradation of EPSs and biogas yield improvement by the addition of hydrolytic enzymes (Liew *et al.* 2020). In the literature, four methods of enzyme addition have been reported (Brémond *et al.* 2018), which include a dedicated vessel for enzyme pre-treatment, direct addition of enzyme to a stage digester, a two-stage process of direct addition of enzyme to hydrolysis and acidification reactor, and addition to recirculating leachate of AD (Brémond *et al.* 2018). Various factors need to be assessed and optimized for effective enzymatic pre-treatment. Along with temperature and pH, these factors include stability, quantity, specificity, and enzyme activity (Divya *et al.* 2015). Generally, the sludge from WWTPs comprises proteins, carbohydrates, and a slight amount of lipids. WAS mainly consists of EPS, which is not readily biodegradable (Brémond *et al.* 2018). Thus, proteases, carbohydrases, and lipases are applied for pre-treatments (Divya *et al.* 2015; Meegoda *et al.* 2018). Enzymes such as glucosidase, glycosidase, amylase, and protease can improve biogas yield by enhancing sludge biodegradability during AD. Application of protease pre-treatment using *Bacillus licheniformis improved* biogas yield by 26% (Bonilla *et al.* 2018). For pre-treatment of WAS, effects of lysozyme, protease, and α-amylase were observed, and lysozyme was the most effective of all the enzymes for hydrolysis and biodegradability of WAS (Odnell *et al.* 2016; Chen *et al.* 2018). The disintegration of sludge flocs and sCOD improvement was 2.23 and 2.15 fold by lysozymes compared to protease and α-amylase (Chen *et al.* 2018). All the aforementioned studies highlight the potential of enzymatic pre-treatment to enhance the performance of AD. However, further research is needed to identify specific enzymes suitable for SS pre-treatment.

3.5. Analysis of pre-treatment techniques

The increments in soluble chemical oxygen demand (sCOD) resulting from various pre-treatment methods applied to SS are shown in Figure 1(a). Among the techniques evaluated, thermal pre-treatment and ozonation emerged as the most effective. Thermal pre-treatment utilizes high temperatures and pressures to break down the structure of SS, promoting increased solubilization (Ngo *et al.* 2021). This method benefits significantly from higher solid contents in the sludge, which allows for more concentrated energy application on the organic components, thus enhancing the breakdown and solubilization of matter

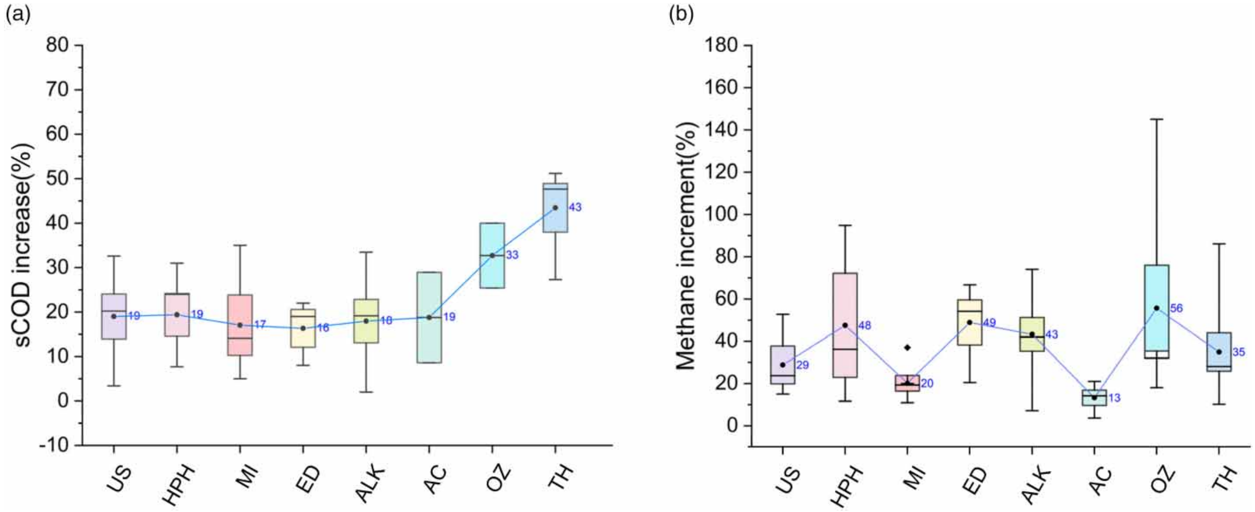


Figure 1 | Enhancement rates in (a) sCOD, and (b) methane yield after different pre-treatment. #Data used for comparison are derived from the Tables S1–S3. US, ultrasonication; HPH, high-pressure homogenization; MI, microwave irradiation; ED, electrical disintegration; ALK, alkali treatment; AC, acid treatment; OZ, ozonation; TH, thermal pre-treatment.

(Chen *et al.* 2020a). Conversely, ozonation utilizes the strong oxidative potential to break down organic materials. The efficiency of ozonation is primarily due to its capability to oxidize OM comprehensively, making it a highly effective pre-treatment method (Catenacci *et al.* 2022b). The observed differences in efficiencies between alkali and acid treatments are rooted in their distinct chemical mechanisms. Alkali treatments facilitate solubilization by breaking down organic structures and increasing the release of proteins and polysaccharides. These components are significantly more soluble under alkaline conditions, enhancing the overall treatment effectiveness. In comparison, acid treatments, though effective, typically achieve lower solubilization because the acidic environment is less disruptive to the sludge (Wang *et al.* 2020). While some treatments, like MI and ultrasonication, show high efficiencies, they may also involve higher operational and capital costs (Cano *et al.* 2015). This economic factor necessitates a balanced approach, combining different treatments to achieve cost-effective solutions without compromising efficiency.

The main goal of sludge pre-treatment is to enhance the efficiency of AD primarily by increasing biogas yield with methane increment as a measure of AD efficiency (Toutian *et al.* 2021). As per Figure 1(b), various pre-treatment methods contribute differently to methane production. Ozonation (56 ± 49%) and electro-kinetic disintegration treatment (49 ± 21%) were the most effective methods for increasing methane yield, followed by HPH (48 ± 42%). Alkali treatment (43 ± 21%) and thermal treatment (35 ± 25%) showed lower increases, while acid treatment displayed the least improvement (13 ± 7%). Ozonation stands out with a 56 ± 49% increase, benefitting from the oxygen released during the process, positively affecting microorganisms and enzymes. Among physical pre-treatment methods, electro-kinetic disintegration proved to be the most effective, significantly boosting methane production by 49 ± 21%.

These variations in methane production underscore the necessity of selecting appropriate pre-treatment methods based on sludge characteristics and desired AD outcomes. Ozonation stands out among all methods for its effective sludge solubilization and biogas enhancement, but it suffers from high operational energy requirements. The efficacy of alkali treatments in enhancing biogas yield suggests that conditions that facilitate the breakdown of complex organic structures into more biodegradable forms can markedly improve AD performance (Tuncay *et al.* 2022). Chemical pre-treatments, particularly alkali and acid methods, although energy-efficient and effective in sludge degradation, use corrosive chemicals that can create by-products inhibiting the AD process and potentially damage equipment. In the case of chemical treatment, the cost is related to the application of acids, alkalis, and other reagents, which are stated to be relatively higher (Hodaei *et al.* 2021). For instance, as per Lee *et al.* (2019), the treatment of one tonne of sludge with alkali pre-treatment to obtain a pH of 10 and 11 resulted in a negative net cost in the range of $0.1 to $2.0 USD. Biological treatment of sludge is considered economical and energetically feasible (Kavitha *et al.* 2014; Banu *et al.* 2018); however, some energy is required for stirring and mixing substrate. Given the cost and potential environmental implications of intensive treatments like thermal hydrolysis and ozonation, exploring combinations of treatments could optimize efficiency, mitigate drawbacks, and reduce the overall environmental footprint. Continuous exploration and innovation in pre-treatment technologies are essential to maximize biogas production and enhance the sustainability of SS management. Techniques like low-temperature thermal-alkali treatments offer substantial improvement in biogas production with relatively lower energy demand and cost compared to more energy-intensive methods, such as ultrasound, ozonation, HPH, electric pulses, and high-temperature thermal treatment (Mancuso *et al.* 2019; Xiao *et al.* 2020). Mechanical pre-treatments are advantageous in existing WWTP due to their compact equipment needs, offering high solubilization rates and improved digestate properties without producing inhibitory by-products. However, they can be costly in terms of full-scale energy use, which can render them uneconomical (Cano *et al.* 2015). Çelebi *et al.* (2021) stated that ultrasonic pre-treatment significantly improved the sCOD concentration of WAS. With a specific energy input of 12,930 kJ/kg TS, methane production increased by 32% compared to the control. However, despite this enhancement, a negative energy balance was reported with a suggestion of implementing partial stream sonication in a full-scale system to improve the overall energy balance. For instance, the combination of sono-thermal pre-treatment applied at laboratory scale for sludge displayed non-viability of sonication due to high energy demand and economic loss. Similarly, for microwave pre-treatment, experiments conducted at a laboratory scale for the analysis of energy balance evidenced the energy intensiveness of the process. For the treatment of one tonne of sludge, the net energy production was negative 466.02 kWh (Rajesh Banu *et al.* 2018). Ultrasonication can be economically viable if the energy requirement at full-scale is < 6 kWh/m^3 (Cano *et al.* 2015). In the case of thermal pre-treatment, the improvement in biogas production and the application of heat exchangers may minimize the pre-treatment cost (Yang *et al.* 2010). Liu *et al.* (2021) achieved a significant outcome by recovering 85% of heat during thermal pre-treatment, demonstrating an effective approach for substantial energy savings. Biological pre-treatments, including enzymatic processes, are the most economical in terms of energy and capital costs. They effectively enhance sludge solubilization and biogas production but are time-consuming and require further development for efficient enzymatic hydrolysis (Daverey *et al.* 2019). The selection and production of hydrolytic enzymes also pose challenges due to their high costs. In a biological treatment, enzymatic hydrolysis can become a feasible option after further research (Brémond *et al.* 2018). Ultimately, the choice of pre-treatment technology depends on a balance between economic feasibility and the specific characteristics of the sludge. While some pre-treatments may lead to greater biogas yields, they may also incur higher operational and maintenance costs (Table 1). Current research and development are focused on integrating various mechanical, chemical, and thermal treatments, with some technologies already implemented at full-scale. For economic analysis along with net profit, other costs, such as treatment, mixing, pumping, labor, collection, tax, transport, digestate, and disposal costs must also be included (Godvin Sharmila *et al.* 2015; Eswari *et al.* 2017; Kannah *et al.* 2017). Further techno-economic analysis is essential to scale up these technologies effectively from laboratory to industrial applications, aiming for sustainable and economically viable outcomes.

4. TECHNICAL CHALLENGES AND FUTURE RESEARCH AREAS

Current research on pre-treatment methods prior to AD mainly aims to enhance biogas yield and focuses on laboratory scale applications, often in combination with various methods. The effectiveness of these pre-treatments largely depends on the type of sludge and AD conditions. WAS from extended aeration systems, which has the lowest intrinsic biodegradability, often yields better methane production compared to primary or mixed sludge (Carrère *et al.* 2008). Additionally, HRT in an AD process is directly proportional to the effectiveness of the pre-treatment in improving digestion rates and methane yield. A critical aspect of pre-treatment is its energy requirement, where the energy balance is defined by the difference between energy generated and consumed post–pre-treatment. The use of a combined heat and power (CHP) engine allows for the valorization of biogas by converting it into electricity and heat. Pre-treatments that utilize heat are generally preferred over those that demand high electricity usage due to better energy efficiencies. High TS content in the sludge enhances the energy efficiency of thermal treatments, as more energy is wasted heating sludge with low solid content. Technologies like pulse electric fields and ball milling are less electrically efficient, whereas high-pressure treatments have shown energy efficiency at full-scale (Cano *et al.* 2015). Despite higher operational costs, such pre-treatments can become economically viable when considering sludge's reduced volume and disposal costs. Chemical pre-treatments require careful supervision due to the potential inhibitory effects of chemicals on AD. They are often used in combination with thermal or physical methods to improve methane yields and reduce energy demands (Chen *et al.* 2020a). However, the impacts on digestate quality and chemical waste minimization must also be considered.

Table 1 | Mechanism, status, and feasibility of different pre-treatments

					Economics		
Pre-treatment	**Control parameters**	**Mechanisms**	**Effects**	**Feasibility**	**Capital cost**	**Operational cost**	**Full-scale technologies**
Ultrasonic	Frequency, power density, solids concentrations, application time	Oxidizing effect, hydro-mechanical shear	Particle size reduction Biogas generation improvement by 40–58% Improved sludge dewaterability	Moderate	High	High	Biosonator, Sonix, Iwe. Tec, Smart DMS, Sonolyzer, Hiescher
High-pressure homogenization	Solids concentrations, pressure	Shear force, turbulence, cavitation, pressure gradient	Biogas generation improvement by 43–90% Odor reduction	Moderate	Low	High	MicroSludge™, Crown, Cellruptor
Microwave irradiation	Wavelength, frequency, solids concentrations, temperature, specific energy, application time	Thermal effect	Biogas generation improvement by 20–53% Pathogen elimination VS removal	Low	High	High	Aspal SLUDGE™, Praxair® Lyso™
Electro-kinetic disintegration	Specific energy, application time	High voltage field	Changes in microbial diversity Biogas generation improvement by 30–31%	Moderate	High	High	BioCrack, OpenCEL, PowerMod
Alkali	pH, dosage, application time	Solvation and saponification	Biogas generation improvement by 38–80% Pathogen inhibition Aids in sludge disposal Dewaterability improvement Increase in solubilisation	Moderate	Low	High	
Acid	pH, dosage, application time	Hydrolysis of hemicellulose, lignin breakdown, cellulose dissolution	Biogas generation improvement by 14–24% tCOD and VSS reduction aid in hydrolytic microbes accumulation	Low	High	High	
Ozonation	Dose, pH, application time	Radical formation		Low	High	High	

(*Continued.*)

Table 1 | Continued

Pre-treatment	Control parameters	Mechanisms	Effects	Feasibility	Economics		Full-scale technologies
					Capital cost	Operational cost	
			Biogas generation improvement by 20–200% Pathogen elimination				Aspal SLUDGE™ (Air Liquide), Praxair® Lyso™
Low-temperature thermal <100 °C	Temperature, application time	Organic particle degradation by thermophiles	Pathogen removal - methane production increment by10–100% - VS reduction up to 20–150%	High	High	Low	CambiTHP™, Turbotec, Biorefinex
High-temperature thermal >100 °C	Temperature, application time, pressure	Cell-wall disruption and protein release	Complete pathogen removal without reactivation Protein degradation Improvement in methane production by 10–150% 10–160% reduction in VS	Moderate	High	High	Lysotherm, Biothelys®, Exelys, Aqualysis, ACH, teH4 +
Temperature-phased anaerobic digestion	Temperature, application time and pH	Hydrolysis and acidogenesis in thermophilic stage, acetogenesis, and methanogenesis in mesophilic stage	Improvement in methane generation by 20–50% 10–70% reduction in VS Improved sludge rheology	Moderate	Low	High	Laboratory scale and full scale
Enzymatic	pH, temperature, enzyme concentration, application time		Improved solubilisation Improvement in methane yield by 12–40% 16–55% reduction in VS	Moderate	High	Low	In few food industries

Source: Gonzalez *et al.* (2018), Elalami *et al.* (2019), Atelge *et al.* (2020) and Khanh Nguyen *et al.* (2021).

Assessing pre-treatment methods based solely on energy feasibility is insufficient. Sustainability evaluations should include technical, environmental, economic, and social factors (Mainardis *et al.* 2021; Balasundaram *et al.* 2022). Recently, Mainardis *et al.* (2021) offered valuable insights into the environmental impacts of various pre-treatment techniques for SS, underscoring the significance of sludge composition in determining the most sustainable approach. These results displayed that the optimal pre-treatment method depends on the specific conditions and scale of operation, highlighting a complex decision matrix for environmental sustainability in SS management. Similarly, Balasundaram *et al.* (2022) provided a comprehensive review of various sludge pre-treatment methods, assessing their energy efficiencies and environmental impacts. Thermal pre-treatment at a temperature below 100 °C had a positive energy balance with lower global warming potential (GWP). On the contrary, thermal pre-treatments above 100 °C involved significant energy use, rendering them less favorable due to their intensive energy requirements and higher GWP. These findings suggested a nuanced approach to selecting and implementing sludge pre-treatment technologies, where operational scale and specific conditions influence the sustainability and effectiveness of each method.

Pre-treatments can also impact the AD microbial community, OM stability, and nutrient content in the digestate, potentially introducing by-products that adversely affect soil health (Solé-Bundó *et al.* 2017; Yang *et al.* 2017). Knowledge gaps exist in how sludge characteristics and structure affect biodegradability. Current literature has not adequately explored the microstructural properties of WAS and their effects on AD. Additionally, the role of sludge constituents, such as heavy metals and their interactions with extracellular polymeric substances (EPSs), in influencing AD efficiency needs further investigation (Liew *et al.* 2020; Khanh Nguyen *et al.* 2021; Toutian *et al.* 2021).

Future research should focus on the role of anaerobic microbes and the impact of WAS structure and characteristics. Studies should use modeling to optimize operational parameters for different sludges in anaerobic digesters and assess the effects of additives on AD. Research should also examine the microstructure of sludge, considering both organic and inorganic components involved in bioconversion during AD. Establishing relationships between the physico-chemical characteristics and microstructure of sludge with anaerobic bioconversion is crucial. The research should aim to enhance AD efficiency by reducing interactions between sludge components, thereby addressing the low degradability issues caused by these interactions.

Incorporating environmental impact assessments through life cycle assessment (LCA) is also crucial when selecting pre-treatment solutions. The LCA approach helps to identify the environmental trade-offs of different pre-treatment technologies. For example, while some methods like ultrasonication may require less energy, they might still lead to significant environmental burdens due to high power consumption at the laboratory scale. On the other hand, methods like thermal hydrolysis, though energy-intensive, can be optimized to utilize waste heat, thereby reducing overall greenhouse gas emissions and improving energy efficiency. Moreover, the choice of pre-treatment can have downstream effects on the properties of the resulting sludge. For instance, certain chemical pre-treatments might enhance biogas yields but also lead to the formation of inhibitory compounds that can affect subsequent AD stages. The application of LCA offers a balanced approach to environmental impact, economic viability, and technical feasibility, leading to more sustainable wastewater management practices.

5. CONCLUSION

Different pre-treatment methods have varied effects on sludge solubilization and the energy efficiency of AD. Physical, chemical, and biological pre-treatments demonstrate improvements in sludge solubilization performance and AD efficiency. Among these, ozonation, electro-kinetic disintegration, and HPH have higher efficiency for energy recovery due to their ability to break down complex organic structures. Alkali treatments, while effective in enhancing sludge solubilization and methane production, involve the use of corrosive chemicals that can create inhibitory by-products and damage equipment. For instance, while ozonation can achieve significant sCOD solubilization and methane production; however, the high energy demands and associated costs pose challenges for widespread adoption. On the other hand, although effective, physical pre-treatments, such as HPH are similarly hindered by their higher energy consumption. Biological pre-treatments, including enzymatic processes, offer a more sustainable and cost-effective approach. These methods enhance sludge solubilization and biogas production with lower energy demands. However, they are often time-consuming and require further development to optimize enzymatic hydrolysis efficiency. The selection and production of hydrolytic enzymes also pose challenges due to their high costs. However, these methods are time-consuming, thus making them less attractive for industrial-scale applications.

Furthermore, the practical application of all these pre-treatment methods is often constrained by their higher energy demands. While pre-treatment methods significantly enhance the efficiency of AD process, careful consideration of their economic and environmental impacts is crucial. Sustainable and cost-effective solutions should be prioritized as these approaches not only improve biogas yields and reduce the environmental footprint but also ensure the long-term viability of SS management. Incorporating standardized analysis protocols to systematically determine energy effectiveness and integrate LCA into the evaluation process will ensure balanced environmental, economic, and technical feasibility.

AUTHOR CONTRIBUTION

V. P. conceptualized the whole article, rendered support in writing collection of information from literature, analysis, compilation and data interpretation, and rendered support in draft writing and editing the article. **Dr S. K.** reviewed the article and edited the article. **Dr B. R. Y.** supervised the article.

AVAILABILITY OF DATA AND MATERIALS

The authors confirm that the data supporting the findings of this study are available within the article.

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DATA AVAILABILITY STATEMENT

All relevant data are included in the paper or its Supplementary Information.

CONFLICT OF INTEREST

The authors declare there is no conflict.

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