

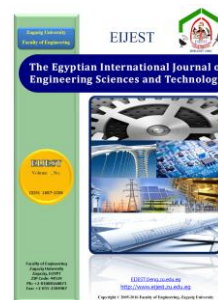


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Sludge disintegration techniques: In-depth review

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ABSTRACT

Nowadays, sludge is considered a concern problem due to the huge quantities of sludge produced from wastewater treatment plants (WWPT), it threatens human health and the environment. Several processes can be utilized to safely remove sludge and utilize it in various applications. Among these processes, anaerobic digestion (AD) is competent, cheap, promising and sustainable process. However, AD is long-time consumer especially for hydrolyses stage because of some complex components like EPS. Hence, to overcome this problem, several sludge pretreatment technologies can be employed before AD, including physical methods such as cavitation, thermal pretreatment and microwaves, chemical processes such as Fenton oxidation and alkaline pretreatment, and biological treatments play a critical role in enhancing AD efficiency besides, the combination of these approaches. These methods have the advantages of sludge disintegration, cell wall and EPS damage, release of sludge to soluble phase and improvement of its dewaterability, resulting in increasing hydrolyses rate, reducing the digestion time (HRT), enhancing AD performance and this leads to more biogas generation which can be utilized as energy and electricity provider to balance the operation and maintenance cost of WWPT or sludge treatment cost. This review paper will focus on the efficient role of different pretreatment methods on sludge properties and AD efficiency.

1 Introduction

Wastewater treatment plant purposes are removing of contaminants and pollutants from sewage, maintaining water quality, and safeguarding the integrity of water bodies. The guidelines for treated sewage flow into water bodies are defined by environmental legislation based on standards of water quality [1]. The activated sludge system is the primary treatment process in more than 90% of municipal WWTPs. In both industrial and municipal wastewater treatment plants, various treatment systems eliminate biodegradable compounds, ,

while physio-chemical processes such as coagulation-flocculation and filtration help separate inorganic and organic particulate matter. This process leads to the production of significant amounts of primary and secondary waste activated sludge (WAS) [2]. Consequently, wastewater treatment generates a considerable volume of sludge, ranging from 5% to 30% of the total wastewater volume [3].

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Sewage sludge is categorised by existence of organic and solid compounds, filamentous bacteria, microbial aggregates, nutrients, pathogens, heavy metals and extracellular polymeric substances (EPSs) [4]. Where, the moisture content of sludge is as high as 98 %, and EPS accounts for 80 % of the WAS mass. EPS influences on the sludge properties including floc stability, surface charge, rheological behaviour, and dewatering. This brings great difficulty to resource utilization and sludge processing [5]. With industry and population expansion, the continuous rise of excess sludge production and its safe disposal and reuse increased the sludge management cost, that can account for as much as 60% of a WWTP's overall operating cost [2].

Agricultural utilize, composting, anaerobic digestion, landfills, recycling as building material, and incineration are the primary methods for disposing of sewage sludge (SS). Each of these management techniques has practical limitations pertaining to both human and environmental health. AD is a cost-effective, environmentally responsible, and efficient treatment method. On the other hand, AD can produce methane gas, which is regarded as a renewable bioenergy source, stabilize sludge, and aid in the removal of pathogens and odor [4].

An important part of the renewable energy sector is the production of biomasses such as biogas, bioethanol, biodiesel, and others during AD. Biogas production is considered a beneficial feature that can partially offset waste management expenses. Nowadays, biogas is produced mostly via SS

digestion which consists of approximately 25–50% carbon dioxide and 50–75% methane, where the content of methane can be utilized as energy, electricity, and heat source [4].

Anaerobic digestion is a complex biochemical process involving microorganisms that form syntrophic consortia. It comprises four stages: hydrolysis, acidogenesis, acetogenesis, and methanogenesis. Hydrolysis involves breaking down large molecular compounds into monomers and smaller molecules. This step is often seen as a limiting factor due to the existence of particulate organic substance and complex macromolecules, such as EPS, produced by microorganisms during the biological treatment of wastewater [6]. Moreover, the presence of heavy metals, inorganic matter, refractory organic matters and emerging micropollutants reduces the functional microorganism's activity, thereby weakening AD performance. In addition to AD faces intrinsic technical challenges, including lengthy operational cycles and stringent working conditions [7], as shown in Fig. 1 [8]. Furthermore, numerous studies have detected that the existence of recalcitrant cell walls, difficult floc structure (EPS) and high molecular weight organic substance obstructs AD hydrolysis. This drawback results in long retention time, needs to a bigger bioreactor, also produces a minor biogas production. Therefore, various studies have suggested sludge pretreatment before AD to enhance hydrolysis, accelerate methane production [4] and disintegrate sludge as well as overcoming the limitations found through hydrolysis and improve

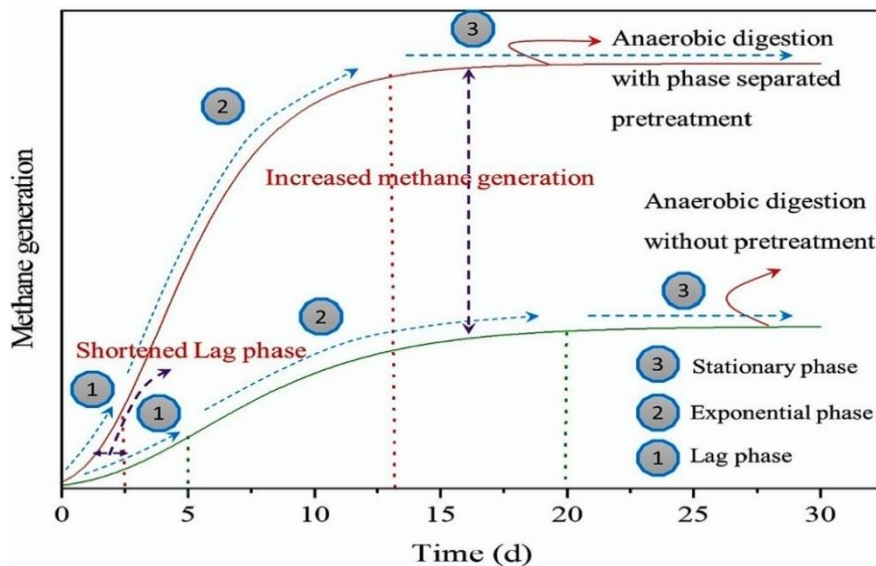


Fig. 1. Profile variation of anaerobic digestion during phase separated pretreatment.

bioconversion efficiency of sludge [9].

So, many different pretreatment/disintegration methods are used to improve AD e.g., physical, chemical, and biological or their different combinations [10]. The main purpose of this paper is to give in-depth review of these methods.

2 Different pretreatment methods

There are several pretreatments methods which used to disintegrate and solubilize sludge. These methods are based on chemical, physical and biological methods, or a combination of these approaches [6], as shown in Fig. 2. Where, these pretreatments before AD process, is necessary to overcome the limitation confronted through AD hydrolysis. They can disintegrate the complex sludge structure, reduce solid mass, rupture complex EPSs, promote SS solubilization, boost substrate solubility and speeds up the biodegradation of organic solid waste during AD. Furthermore, these pretreatment processes increase biodegradability and methane production [4].

2.1 Physical pretreatment

Physical/mechanical methods for substrate's pretreatment are a decrease in particle size through applying of a physical force. Especially, this changes the organic components into minor and more soluble fractions [9], and thus raises the particle surface area to augment AD [4]. Moreover, it has benefits of no chemical substances are utilized, nor are the functional microbes augmented to enhance the bioconversion efficiency. Most of these techniques are energy intensive methods, that could convert in

event of extreme energy recovery for pretreatment electricity or heat requirements, resulting in energetically self-sufficient procedures [9]. Physical pretreatment can be divided to mechanical pretreatment and thermal pretreatment methods.

2.1.1 Mechanical pretreatment

2.1.1.1 Cavitation

Cavitation is the term for the process that occurs when vapor or gas-filled cavities form, grow, and collapse in a small time (usually milliseconds) [11][12]. Cavitation is the conversion from liquid to vapor phase, it occurs when local liquid pressure reaches saturation value at a certain temperature. Thus, cavitation process has been applied in several fields including biodiesel synthesis, cell disruption, disinfection, sludge treatment, polymer degradation, and the breakdown of various organic compounds like pesticides, textile dyes, phenolics, and pharmaceutical residues [11].

Cavitation creates localized conditions of extreme temperatures (ranging from 500 to 15000 K) and pressures (between 100 and 5000 atmospheres), yet the overall environment stills at atmospheric conditions [13]. Additionally, the great temperature and pressure generated during the cavitation, the cavitation bubbles collapse can produce a range of physicochemical effects resulting in various physical and chemical changes [11]. Physical effects of 550 MPa shock wave is produced at a speed equals 2000 m/s, a micro jet creates a 450 MPa water hammer at 100 m/s [14] and a high shear wall stress up to 3.5 kPa [12], resulting in particles disintegration and

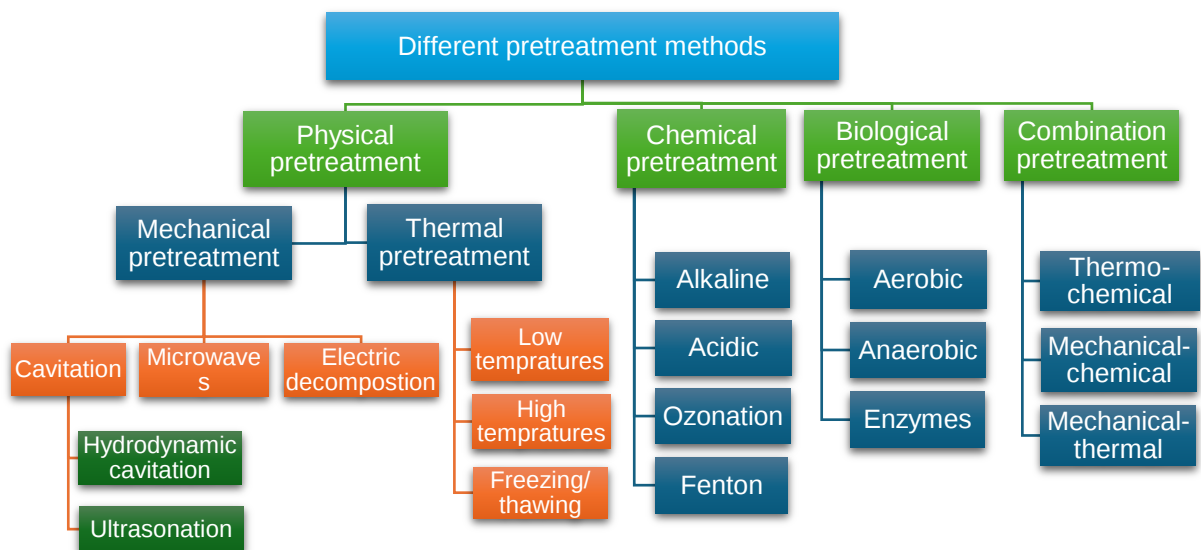


Fig. 2. Different sludge pretreatment methods.

microorganisms lysis causing biogas production improvement through biomass anaerobic digestion [14]. While, for the thermal effect, bubble collapse produces 2000 – 6000 K local hot spot and causes heat transfer of 1010 K/s within one microsecond, this process improves the dewaterability of sludge and decreases its viscosity by inducing cell lysis and compromising the integrity of cell walls [14]. On other hand, for chemical impact, under extreme cavitation conditions and energy produced by bubble collapse, water molecules can be decomposed into a various species with a very oxidation potential, containing hydroxyl radicals: (H·), (OH·), (OOH·) and H₂O₂, that react with organic compounds in the wastewater or treat sludge, thus speed up chemical reactions and deactivate and eliminate microorganisms during hydrogen bonds decomposition [11][14].

Cavitation can be categorized into four types based on how it is generated: acoustic cavitation (AC), hydrodynamic cavitation (HC), optical cavitation (OC), and particle cavitation (PC) [11][12]. The AC is caused from pressure variations in the liquid through sound waves, and HC is created by alterations in pressure and flow through special constructions. On the other hand, in optic cavitation, the light of a pulsed laser converges in a liquid causing the local deposit of energy and then transient bubbles are generated as a result of optical breakdown. While, particle cavitation is formed by any other kinds of elementary particle beam, e.g. a proton rupturing a liquid in a bubble chamber. Nevertheless, both OC and PC are utilized for single bubble cavitation and not appropriate for creating enough intensities for physical and chemical applications [11], where the most common types of cavitation are acoustic and hydrodynamic [15]. As illustrated in Fig. 3 [3], the bubbles of hydrodynamic cavitation have behavior patterns like the bubbles of

acoustic cavitation [16].

A. Hydrodynamic cavitation

Hydrodynamic cavitation (HC) is a promising technique for process intensification, because of cost-effective operation, its high energy efficiency and ability to induce chemical reactions in addition to scalability [16]. Since, HC is produced by changes in pressure and flow, that are frequently brought about by certain constructions (nozzles, venture tubes, and orifice plates), mechanical rotation of rotating-type devices, and vortex-based devices [11] as presented in Fig. 4 [3], or high-pressure homogenizers, all those dynamic devices are high shear stresses that act a significant role in breaking up WAS flocs [17].

Based on the equation of Bernoulli, the liquid pressure reduces with kinetic energy growth. So, once local pressure falls to threshold pressure for cavitation under saturated vapor pressure at the working temperature, cavities start to create and grow. Based on flow conditions, the produced cavitation bubbles size often differs from a few nanometers to a few millimeters. When the pressure reclaims or increases over vapor pressure, the cavities failure, and the micro-jets create turbulence downstream of constriction [11] as illustrated in Fig. 3 [3]. Therefore, hydrodynamic cavitation intensity is based on the intensity of turbulence and cavities number created. Where, the turbulence intensity is associated with constructions geometry and conditions of liquid flow. Cavitation number (C_v) is a dimensionless number, which describes HC condition. It is given by the next Eq. (1):

$$C_v = (P_2 - P_v) / (.5 \rho V_o^2) \quad (1)$$

Where: P_2 is wholly reclaimed downstream pressure, P_v is liquid vapor pressure, V_o is throat velocity of cavitation device, and ρ is liquid density. C_v reduces by the increase in working inlet pressure for each

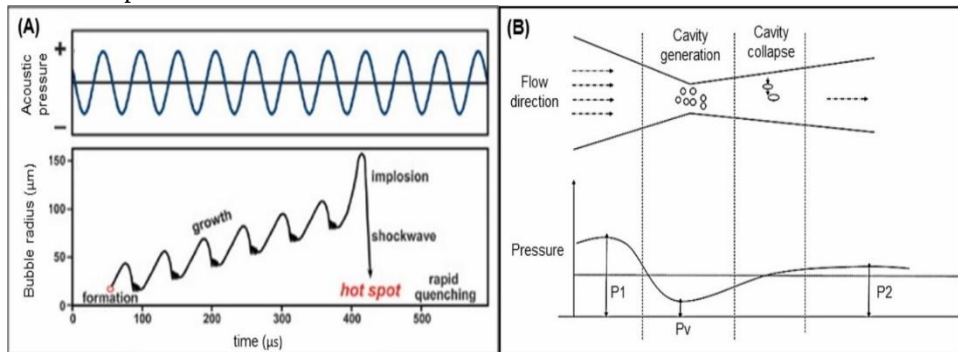


Fig. 3. Cavity generation and collapse in (A) acoustic cavitation and (B) hydrodynamic cavitation as a function of pressure.

traditional cavitation devices (venturi or orifice devices), where throat velocity of cavitation device rises with the increase of working inlet pressure [11].

cavitation beginning and bubbles creation normally occur at $C_V \leq 1$. Nevertheless, cavitation phenomenon can occur even at C_V greater than 1 as a result of little quantities presence of suspended solids and dissolved gases. Generally, higher number of bubbles can be created at lower values of C_V , even though it causes a reduce in bubbles collapse intensity. Where, the too low values of C_V result in

pressure. The favorite value of C_V , leading to effective pretreatment of biomass and disintegration, is principally between 0.07 to 0.15 [12].

Another shape of HC is high pressure homogenization (HPH), which is a hopeful untraditional mechanical pretreatment technology [18], it utilizes the impacts of extreme pressure, cavitation, turbulence, and strong shear within high pressure homogenizers, causing important changes in sludge characteristics [7]. Furthermore, HPH's performance can be influenced by various parameters

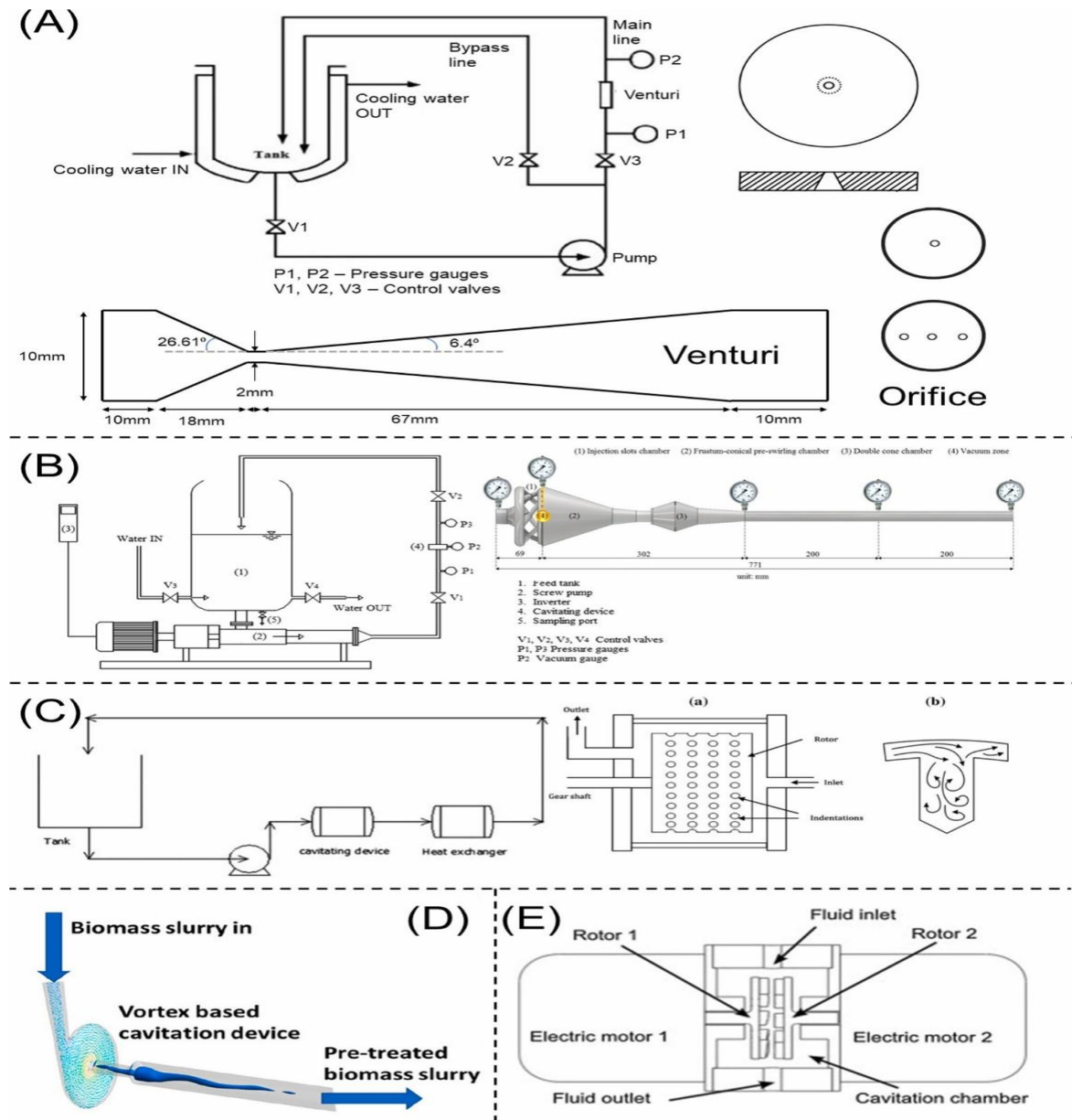


Fig. 4. Several types of HC devices for pretreatment of waste sludge: (A) venturi and orifices, (B) swirling jet cavitation, (C) rotor and stator assembly, (D) vortex-cavitation, and (E) rotation generator. super cavitation leading to a decrease in collapse such as the number of orifices through valve,

pressure, valve properties, and the concentration of suspended solids in the sludge [19]. Through this method, sludge flocs rupture and cell membrane breaks, releasing intracellular matters. Therefore, high pressure homogenization improves biodegradation and sludge disintegration performing

[20]. Additionally, high-pressure homogenization reduces the viscosity of sewage sludge, average particle size, surface tension, and zeta potential, while boosting SCOD, VFAs, and specific surface area [7].

Table 1: Comparison between different types of hydrodynamic cavitation devices [2][11][12][19].

Type	Benefits	Drawbacks
Orifice plate	1. Plain construction 2. resilient design 3. Small power utilisation and maintenance cost 4. Easy to control and work	1. Great probability of erosion and clogging for waste involves solid particles. 2. Extreme loss in pressure and energy disperse
Venturi tube	1. Plain construction 2. Small power utilisation and maintenance cost 3. Maximum residence time 4. Easy operation 5. Smooth converging and diverging sections	1. About higher construction cost than orifice especially slit venturi 2. Great probability of erosion and clogging for waste involves solid particles. 3. Great pressure loss and energy disperse
Vortex based type	1. Decrease the destruction to the surface of cavitation device and pipe 2. Low cost of maintenance and working with respect to rotating kind 3. Low overpressure of the system needed 4. Fitting for waste containing solid particles 5. No require moving parts to vortex creation	1. Difficult scaling-up 2. sensitive to clogging 3. Flexibility of governing the cavitation intensity is much fewer than that of orifice and venturi
Rotating type	1. Fitting for waste involving solid elements 2. Smaller pressure loss 3. Greater mass transmission efficiencies	1. Extreme power utilisation and maintenance cost 2. Difficult construction
High pressure homogenization	1. Short contact time 2. Homogeneous flocs sludge dimension 3. Real-scale applications 4. Easy to employ 5. No odour creation and better sludge dewaterability after AD	1. Mechanical deterioration 2. Weak pathogen removal 3. Extreme energy utilisation 4. Clogging troubles because of coarse in addition fibrous particles 5. Extreme erosion and tensions in pump besides homogenizing valve

B. Ultrasonication

Ultrasonication (US) is a widely researched mechanical pretreatment technique that augments biodegradability of sludge. It creates hydro-mechanical shear forces through cavitation, which helps break down the sludge structure [4]. Since ultrasound is a type of sound wave with a frequency more than person audible one. Whereas in AC, acoustic irradiation is responsible for the creation, expansion, and collapse of microbubbles, the

expansion and compression cycles of ultrasonic waves formed by wave transition into the irradiated medium make positive and negative pressures in liquid, that cause microbubbles to expand and compress, respectively [12] The microbubbles generated in this process collapse violently within little microseconds after attaining a critical size, triggering cavitation. This rapid and intense collapse creates extreme conditions, with local temperatures reaching approximately 5000 K and pressures

exceeding 500 bars [20] as indicated in Fig. 5 [9]. The power density is commonly ranged from 100 to 1000 W/L for crystallization processes in various kinds of AC reactors. In sludge disintegration, power density more than 500 W/L might result in the deactivation of sludge. Furthermore, the optimal ultrasonic power density values were ranged from 2000 to 3000 W/L for the extraction of microbial flocculant from waste activated sludge. While, Frequency inversely affects the size of bubbles and the energy produced due to the bubbles collapse. Therefore, lower frequencies in a range of 20 to 50 kHz, can cause extreme bubbles collapse and the intensification of physical impacts of cavitation [12]. So, ultrasound are typically about 20 KHz. Particle size reduction is very rapidly in the initial 15 min for US. While, at the same power density, other new investigations noted more than twice rise in SCOD within 15–20 min [3].

Sludge solubilization and hydrolysis involve a number of processes that can be triggered by the energy used in the form of ultrasonic waves: (1) hydromechanical shear forces; (2) thermal decomposition of volatile hydrophobic matters; (3) formation of radicals; and (4) increase in temperature [1]. These chemical, physical and biological influences lead to reduce in particle size, higher solubilization of organic compound, enzyme release as well as motivation of biological activity [4]. Moreover, strong shear forces lead to collapse of walls, flocs, and cell membranes, leading to volatile solids hydrolysis growing. Besides, solids content and specific energy, other elements such as concentrations of dissolved gases, viscosity, and pressure can affect cell disintegration degree [1].

2.1.1.2 Microwave irradiation

Microwave (MW) irradiation method is another choice to traditional thermal pretreatment to save energy, raise the volatile solids destruction and destroy pathogen [1]. Microwaves are classified as a type of electromagnetic radiation [9], with

wavelengths in range between 1 mm to 1 m and corresponding oscillation frequencies of 300 MHz to 300 GHz [2][20].

Microbial cells and EPS in sludge are disrupted by thermal and non-thermal impacts (electrical, chemical and magnetic) through the use of microwave pretreatment (MP) [7]. Thermal effects happen from the interaction between the electric field of radiation and dipolar molecules, leading to an rise in temperature as a result of molecular rotation and friction. In contrast, non-thermal effects arise from fast change in dipole orientation of the macromolecules making up the cell membrane, which can break hydrogen bonds between molecules, hence disrupt the membrane. While the thermal effects of microwaves have been extensively researched, non-thermal effects are more challenging to quantify, leaving their overall impact on the pretreatment process not fully understood [6]. Generally, microwave can disrupt sludge's floc, release bound organic substance and destroy cell walls [9].

2.1.1.3 Electric decomposition pretreatment

Electrochemical (EC) method is an efficient technique, involving electro-deposition, electro-coagulation, electro-oxidation and electro-flotation, where electro-oxidation was utilized in wastewater treatment due to efficient degradation of refractory impurities, [21].

Electric decomposition can serve to improve microbial cell membrane permeability and break cell wall, also contribute to release of lysozymes and hydrolases [7]. The advanced electrochemical oxidation depends on the hydroxyl radicals creation at electrode surface that results in cell breakage, conversion of organic matter to low molecular weight compounds and release of intracellular material [22]. Moreover, Platinum, titanium, yttrium oxide, ruthenium oxide, or home-made composite conductive materials are the electrodes that are used, while electric decomposition performance is

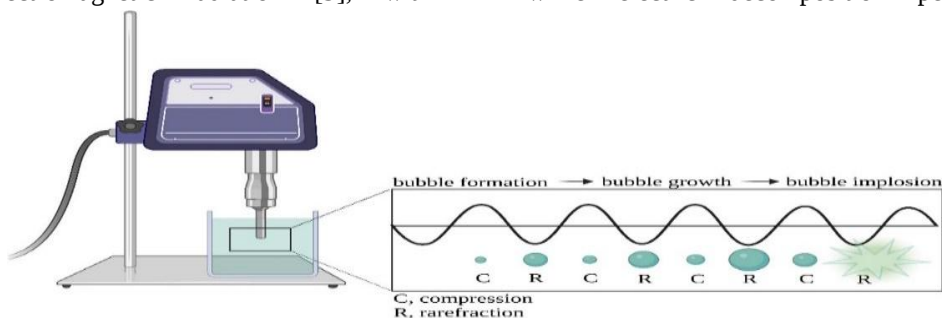


Fig. 5. Cavitation phenomenon created by ultrasonication.

determined by some elements for example voltage, electrolysis duration, electrode distance and current density [7].

There is another technique known as electric pulse that employs high voltage pulsing electric fields (20–30 kV) [9][6], it can disrupt bacterial cell membranes, where the utilized electric field directly

assaults the principal components of bacterial membrane and cell walls, besides, floc structure disruption, releasing intracellular organic matter, and decreasing complex organic molecules to simpler chemical forms[6].

Table 2: Recent researches of mechanical pretreatment methods.

Method	Conditions	Results	Ref.
1. Cavitation: A. Hydrodynamic cavitation:	Orifice: 20 holes of 2 mm in diameter, P = 3 bar, time = 240 min.	SCOD = 319.66 mg/L, DD _{SCOD} = 22.98 %.	[23]
- Simple device: orifice, venturi, nozzle.	Orifice: 27 holes of 1 mm diameter, P= 0.7 MPa, times = 1 to 20 min.	DD values between 11.52 and 23.67%	[24]
	Orifice: 27 holes of 1 mm diameter, difference in pressure between up-stream and downstream = 4.5 bar.	DD increased from 1.4 to 11.9%.	[25]
	Slit venturi: C _v = .2 , P = 3.9 bar, 810 times of passes.	DD = 53.46% for TS = 1.1 %, 67.17% for TS =1.4% and 76.09% for TS =2.1 %.	[26]
	Nozzle: 1.2 mm; P = 12 bars; time = 15 to 90 min.	DD =11% at 15 min, 23% at 30 min, and 32% at 45 min	[27]
- Vortex-based device	Vortex device: pressure drop across HC = 250 kPa, number of passes = (10, 20, 40 and 80 passes).	SCOD raised by more than 25%, BMP more than 80% of theoretical BMP, more than 65% reduction in volatile solids.	[28]
	Vortex cavitation circulating fluidised grinding (VCCFGT): P = 0.30 MPa, filling ratio of 1.6%, time of 60 min.	Mill balling raised SCOD, DD _{SCOD} , protein, and carbohydrate by 218, 229, 177, and 371% respectively.	[29]
	Low-pressure swirling jet cavitation (EW 25): two and four injection slots	DD _{PCOD} reached to 0.96 % in test 2HC, 0.69 % in test 4HC.	[30]
	Swirling jet-induced cavitation: cattle manure, P = 6 to 8 bars.	DD (based on maximum SCOD) raised by 5.8%, 8.9%, and 15.8% for 6, 7, and 8 bar.	[31]
- Rotating device	Power of 4 Kw, 2800 rpm rotation speed.	37% COD solubilization, 42% TOC solubilization, 19.6% increase in CH ₄ yield.	[32]
	Rotational speeds = 1500, 2500, and 300 rpm, energy density E _L = 35 kJ/L..	SCOD as 2.6±0.7-fold at 1500 rpm, 3.1±0.9-fold at 2500 rpm, 3.6±0.9-fold at 3000 rpm.	[33]
	Rotor-stator hydrodynamic cavitation reactor	77.3% in particle size reduction, 500% increase in SCOD.	[34]
	Power = 2.2 Kw; speed = 2800 rpm, E _L = 70, 140, 210 kJ/L.	SCOD increased from 261 to 5810 mg/L, DD _{COD} = 48% at E _L of 210 kJ/L.	[35]
- High pressure homogenisation	HPH with pressure of 40 MPa for pretreated sewage sludge.	SCOD increased from 175 ± 11.35 to 3131 ± 121.02mg/L, DD _{COD} = 24%.	[36]
	Homogenization treatment on sludge with 1 to3 cycles, P = 0 to 60 MPa.	DD _{COD} = 37.87%, zeta potential and surface tension reduced by 79.84% and 14.44% respectively, for 3 cycle and 60 MPa.	[37]

B. Ultrasonication	TS = 9.58 g/L, P = 80 MPa, four cycles.	DD _{COD} = 43.94%.	[38]
	Density = 0.45W/ML, specific energy = 801.58 kJ/g TSS, time = 30 min.	153.84% increase in COD solubilization, SVI reduced by 26%, DD = 27.65%.	[39]
	0.73 W/ML, time = 10 min.	An increase in SCOD of 3.4 fold 32% increase in methane generation, 15% increase in biogas generation.	[40]
	24 kHz, 255 W, time = 8 min, sonication energy input (2,200 to 8,800 kJ/kg dry solid), WAS.	At 8,800 kJ/kg TS, CST raised by nearly 100 times, disintegration degree reached to 8.5%.	[27]
	US densities = 0.04 to 0.1 W/ML, 9690 kJ/kg TS for biological sludge	860% increase in dissolved organic carbon, 340% increase in SCOD, 716% increase in total nitrogen.	[41]
2. Microwaves	A gradient rate = 2 °C/min, T = 90 °C, input power = 900 W.	SCOD increased from 94 to 2452.6 mg/L, disintegration (based on SCOD _{NaOH}) = 70%, (42-45%) increase in methane production.	[42]
	2450 MHz, 600 W in ambient pressure, time = 20 min for sludge (SS) and food waste (FW).	SCOD of SS increased from 4.75 ± 0.50 to 24.58 ± 8.70 g/L and for FW from 38.70 ± 20.65 to 44.20 ± 11.56 g/L.	[43]
	WAS at microwave energy of 336 kJ/kg, 800 W, T = 80 °C for 3.5 min.	SCOD/COD raised from 3.0% to 9.2%, TVFA increased from 196 to 202 mg/L.	[44]
	Thickened waste activated sludge, T = 50–175°C.	SCOD/TCOD increased from 9 ± 1 to 35 ± 1% at 175°C, higher biogas of 31 ± 6% after 18 days for sludge irradiated to 175°C.	[45]
3. Electric decomposition pretreatment	5 g/ L Na ₂ SO ₄ , 3 A of current, pH = 10, time = 100 min.	DD _{NaOH} = 23.93% for TiO ₂ /RuO ₂ electrodes DD _{NaOH} = 14.26% for Pt/TiO ₂ electrodes.	[46]
	Pretreatment voltage = 8 to 15 V, time = 30 min, WAS.	SCOD increased from 29 ± 7 to 350 ± 14, 660 ± 11, 715 ± 21 and 750 ± 21 mg COD/L at 8, 10, 12 and 15 V, respectively.	[47]
	pH = 7, reaction time = 30 min, current density = 150 A/m ² .	DD _{NaOH} = 84.23%, SCOD = 675.2 mg/L.	[48]
	60 V, 83.5 min, 11.6 spacing, petroleum sludge	SCOD solubilization increment by = 230%, VFA solubilization increment by = 172%.	[49]
	Focused pulsed treatment.	Soluble compounds: VFAs, sugar and protein increased by 110, 300 and 460%, SCOD increased by 220%, SCOD/TCOD was 3.3%.	[50]
	High voltage sludge disintegration.	COD in sludge liquor raised from 602 to 6111 mg/L by about 10 times.	[51]
	Pulse-power pretreatment of WAS: voltage = 19 KV, frequency = 110 Hz.	SCOD/TCOD raised from 0.040 to 0.180, 2.5 times higher gas yield than raw sludge.	[52]

2.1.2 Thermal pretreatment

Thermal pretreatment is a widely utilized technique in commercial applications aimed at

improving hydrolysis in anaerobic digestion. Where, thermal pretreatment of SS has applied at various temperatures (50–250 °C), it can be classified as low

temperature (less than 100 °C), high temperature (more than 100 °C) and freezing/thawing pretreatment [4]. Consequently, Thermal pretreatment can break down the chemical bonds for cell walls and allow cellular components to be released into the liquid phase, also enhance water release and result in the biosolids dewaterability [53]. Several studies informed that thermal pretreatment performance depends on applied temperature and

time. Accordingly, temperatures in range of 160 to 180°C and duration from 30 to 60 min are utilized leading to growths of 40–80% in biogas generation, while increases of 20–50% in biogas generation for low temperature at 70 °C and time of hours to numerous days. Consequently, high temperatures pretreatment is more active to improve sludge AD [1].

Table 3: Recent researches of thermal pretreatment methods.

Method	Conditions	Results	Ref.
Low-temperature pretreatment	T = 60–120 °C, saline waste activated sludge.	SCOD raised by 4.2–11.9 times, proteins solubilization by 8.9–35.9 times, carbohydrate solubilization by 5.6–43.8 times.	[54]
	T = 60 °C, pH = 10, time = 24 h, WAS.	Maximum methane yield = 215.6 mL/g COD which was 46.6% higher than untreated WAS.	[55]
	T = 50 to 80 °C, time = 15 to 90 min, sewage sludge.	After 30 min of thermal pretreatment at 80 °C: DD _{COD} was 16.6%, biogas production raised by 24.4% (20 days).	[56]
	T = 80, 100, and 120°C, time = 60 min.	Increase of SCOD = (245.26 - 819.79%), COD solubilization = (2.38 - 7.46%).	[57]
	T = 70 to 90 °C, time = 1 to 15 h.	Disintegration rate increased from 15% to 25%, 15% to 28% and 19% to 30% at T = 70, 80 and 90 °C, respectively.	[58]
	T = 70 to 90°C time = 24 h.	COD solubilization raised from 4.5% to 29.6%, 30.3%, 34.8%, and 41.1% at 60, 70, 80 and 90 °C respectively.	[59]
High-temperature pretreatment	T = 100 to 200 °C, time = 60 to 240 min, mixed sludge.	Ratio of COD solubilization raised from 11.2% to 15.1%, and 25.1% after the time raised from 60 to 120 and 240 min. respectively at 175 °C.	[60]
	T = 180 °C.	Solubilization of the digestate reached to 42.1%.	[61]
	T = 170 °C, time = 30 min.	COD solubilization degree =28%, 35% for carbohydrate, while 49% for protein, VSS reduction = 41%, TSS reduction = 34%.	[62]
	T = 160 °C, P = 0.6 MPa, time = 30 min.	SCOD increased from 270 to 11,350 mg/L, with a 42-fold increase.	[63]
	T = 110 to 150 °C, time = 20 min.	DD raised from 14% at 110 °C to 20% at 150 °C	[24]
	Thermal hydrolysis pretreatment of sewage sludge.	SCOD/TCOD = 29.7 ± 0.2%, TVFAs reached 8.2 ± 0.1 g/L, S-protein / T-protein increased from 43.3 ± 0.8% to 67.1 ± 0.6%.	[64]
	T = 165 °C, time = 50 min.	SCOD/COD ratio was 45.21%.	[65]

	T = 180°C, TS = 11%.	Maximum biogas = 3645 ml with an enhancement of 60.57%.	[66]
	T = 134 ± 1 °C, P = 3.4 bar, time = 30 min.	DD = 23.64 ± 0.23%.	[67]
Freezing/thawing	Freezing/thawing gravitational surplus activated sludge using dry ice; volume ratio of surplus sludge to dry ice of 1:1.	SCOD increased from 63 to 840 mg O ₂ /dm ³ , supernatant turbidity raised from 10 to 101 NTU, SVI was reduced by 56%.	[68]
	Freezing: - 18 °C for 72 h, and thawing: 29°C for 3 h, combined with <i>Geobacillus</i> sp. G1 (60 °C for 6 h, 10 % <i>Geobacillus</i> sp. G1 adding after freezing/thawing).	Maximal soluble protein changed from 40 ± 6 to 1226 ± 24 mg COD/L, accumulated short-chain fatty acids increased from 248 ± 81 to 3032 ± 53 mg COD/L for combined pretreatment.	[69]
	Freezing: - 25 °C for 24 h, thawing: 20 °C for 12 h.	SCOD increased from 3190 to 6756 mg/L, VFA increased from 1757 to 3755 mg/L.	[70]

2.2 Chemical pretreatment

Chemical pretreatments (CP) contain the chemical substances utilized for hydrolyzing sludge. Whereas a variety of chemical processes, including ozonation, acid and alkaline hydrolysis besides other oxidation techniques like Fenton reaction, have been studied [6]. The primary aims of chemical pretreatment techniques are sludge's floc structure disintegration with the help of strong chemical matters [9].

Alkaline pretreatment is acceptable for sludge solubilization, while acid pretreatment is acceptable for lignocellulosic biomass [1]. Acid or base addition can also be carried out at ambient or moderate temperatures avoiding the high temperature need [20]. Despite these benefits, which are correlated to simple equipment and ease of CP operation, these techniques are not favored as physical ones, because of chemicals purchase that increases the cost of operation and maintenance [9].

2.2.1 Alkaline pretreatment

Alkaline pre-treatment dissolves the cell membranes and release the intracellular matter from the cells in solution, that is accessible for the fermentation process [10][53]. Furthermore, it has a powerful effect on cellular matter damage and EPS solubilization, and it significantly speeds up the solubilization and hydrolysis of residual sludge, specifically WAS [6]. NaOH has the highest efficiency in sludge solubilization and improving biogas yield among alkaline reagents with a ranking of efficiency being NaOH > KOH > Mg(OH)₂ and Ca(OH)₂ [6][20].

2.2.2 Acidic pretreatment

Acidic pretreatment has less attention than alkaline pretreatment for SS AD. Conversely, this pretreatment is highly active for treating lignocellulosic matters existing in SS as it helps hydrolytic microbe aggregation under acidic conditions and lignin breakdown [4]. Acid pre-treatment destroys polymers into monomers or oligomers, that raises the rate of digestion [6]. Whereas the most famous chemical reagents utilized in acid pretreatment include HCl, H₂SO₄, H₃PO₄, and HNO₂, while pH varies in value from 1 to 5.5 [9].

2.2.3 Ozonation

Ozone gas has a great oxidation potential on different organic compounds. It can be used as a pretreat for sludge to facilitate the partial oxidation and hydrolysis of biodegradable organic materials, resulting in increased biogas production. For sludge solubilization, the ideal ozone dose is ranged between 0.05 and 0.5 g O₃/g TS based on sludge properties and applied conditions of pretreatment. However, the ozonation efficiency is associated with ozone reaction kinetics and mass transfer, where the kinetic reactions between sludge and soluble ozone and have a minor average rate. Therefore, more solubilization after a specific dosage, cannot be noted. Moreover, partial or full mineralization of the released cellular materials can be created at very high dosage, affecting on final methane productivity [1].

The mechanism by which ozone acts involves its rapid breakdown into radicals that oxidize both soluble and particulate organic substances through direct and indirect reactions. The rate of direct reactions is influenced primarily by the structure of the reactants, while indirect reactions rely on the high

reactivity of hydroxyl radicals, which do not target specific compounds. Ozone decomposes organic matter in two stages: first, solubilization occurs due to the disintegration of suspended solids, and mineralization through the oxidation of soluble organic substance. The goal of the pretreatment process is to achieve partial oxidation and sludge hydrolysis, avoiding complete oxidation [6].

2.2.4 Fenton Oxidation

Hydrogen peroxide (H_2O_2) is a popular oxidant and commonly used in the advanced oxidation process (AOP), it can produce potent oxidative hydroxyl radicals ($OH^\cdot$) that act a significant role in water treatment e.g. it can deactivate bacteria through the disinfection procedure as well as release water by oxidizing EPS through sludge dewatering. Moreover, $OH^\cdot$ can damage cell walls for microorganisms or bacteria, improve the penetrability of cell membrane, and break the cell walls in order to release intracellular water from cells [71].

Fenton process is another widely recognized oxidation method which contains hydrogen peroxide (H_2O_2) reactions with iron ions (Fe^{+2}) catalyst to

create very energetic hydroxyl radicals ($OH^\cdot$). that have a high-level oxidation potential of +2.80 V (in acidic conditions) than hydrogen peroxide (+1.36 V), and ozone (+2.07 V) and are especially efficient in EPS disintegration and cell lysis of microorganisms in sludge, causing in intracellular substances release. Besides, it has been intensively applied for enhancing sludge dewatering [20]. Additionally, advanced oxidation processes break down the complicated structures that shield microbes from reactions, resulting in substrate solubilization and improved biodegradability [53]. On other hand, pH, temperature, reaction time, hydrogen peroxide concentration and Fe^{+2} concentration are the important parameters which influence on pretreatment efficiency [1].

Table 4: Recent researches of chemical pretreatment methods.

Method	Conditions	Results	Ref.
Alkaline pretreatment	Dose:20, 40, 60 and 80 mg NaOH/ g TS of dewatered activated sludge.	Solubilization = 1.83 to 10.36%.	[57]
	pH = 12 (NaOH).	Dissolved organic carbon, TDS, proteins, and carbohydrates increased from 0.9 ± 0.1 , 48 ± 4 , 2.8 ± 0.17 and 1.2 ± 0.1 mg/g TS to 13.7 ± 0.7 , 501 ± 6 , 131.1 ± 1.94 and 23.7 ± 0.7 mg/g TS, respectively.	[72]
	pH = 8 to 11 using: NaOH and Na_2CO_3 .	At pH 11, DD % = 16.2% for NaOH, 10.6% for Na_2CO_3 .	[25]
	Dose: 0.1 mol/L of NaOH	Disintegration degree = 26.9%, organic degradation rate = 38.3%, biogas yield = 0.65 L/g VSS.	[73]
Acid pretreatment	pH = 8 to 13 using: NaOH, KOH, and $Ca(OH)_2$.	DD% = 40.10% for pH 13 by NaOH.	[24]
	pH = 2 (HCl).	Dissolved organic carbon, TDS, proteins, and carbohydrates raised from 0.9 ± 0.1 , 48 ± 4 , 2.8 ± 0.17 and 1.2 ± 0.1 mg $\cdot$ gTS $^{-1}$ to 1.2 ± 0.2 , 286 ± 5 , 8.9 ± 0.48 and 4.4 ± 0.2 mg $\cdot$ gTS $^{-1}$, respectively.	[72]
	pH = 2 to 5 using: HCl and H_2SO_4 , WAS.	At pH = 2, DD% = 3.8% by HCl, 1.6% by H_2SO_4 . 24.5% reduction in SS, 39.0 % reduction in VSS,	[25]

Ozonation	Peracetic acid with dosage of 30 mg PAA/g SS, time = 2 h.	530% increase in SCOD, 177% increase in protein, 20% increase in cumulative biogas.	[74]
	pH = 2 by using HCl, WAS.	Methane yield increased by 14.3% (HRT = 12 day and 35 °C).	[75]
	Ozone dose: 0.06 g O ₃ /gTSS.	The highest efficiencies of daily removal = 35% for COD, 32% for TS, 42% for VS, 60% for TSS and 69% for VSS.	[76]
	Ozone with dosages of 0.05 and 0.1g O ₃ /gTS, in short term (10 days) and long term (30 days) sludge retention times.	At 10 days, TS reduced from 38.8 to 32.8 g/L for 0.05 gO ₃ /gTS and to 34.2 g/L for 0.1 gO ₃ /gTS, respectively, biogas production yield increased by 62.23% for 0.05 gO ₃ /gTS and 53.12% for 0.1 gO ₃ /gTS.	[77]
	Ozone dose: 0.1 g O ₃ /kg TS.	Disintegration degree = 51.1%, SS reduction = 34%, increase in methane production = 55%.	[78]
Fenton Oxidation	Classical Fenton process (CFP): Fe ⁺² = 6 g /kgTS, H ₂ O ₂ = 40 g /kgTS at pH = 3 modified Fenton process (FTP): Fe ⁰ = 4 g /kgTS, H ₂ O ₂ = 20 g / kg TS at pH = 3.	Disintegration degree (DD) = 27.12 % for CFP and 24.41 % for FTP.	[79]
	CFP and FTP: catalyst iron dose of 4 g/kg TS, H ₂ O ₂ dose of 40 g/kg TS and pH = 3 for 60 min.	Disintegration efficiencies enhanced to 23.6% in CFP and 16.7% in FTP, relative increase in biogas and methane productions = 30.2% and 38.0% by CFP while 24.4% and 26.8% by FTP.	[80]
	H ₂ O ₂ dose of 60 g /kgTS, 0.07 g Fe ⁺² /g H ₂ O ₂ ratio, 150 rpm for 1 h to acidified sludge.	Suspended solid reduced from 26.98 to 21.27 gSS/L with 21% solubilization, VSS reduced from 19.31 gVSS/L to 14.46 gVSS/L with 25% solubilization.	[81]
	H ₂ O ₂ = 50 g /kg DS, 0.07 g Fe ⁺² /g H ₂ O ₂ , 60 min, pH of 3.	COD and BOD release increased from 421 and 198 mg O ₂ /L to 2507 and 1403 mg O ₂ /L respectively.	[82]

2.3 Biological pretreatment

Biological pretreatments are environmentally friendly approaches that use aerobic, anaerobic, and enzymatic processes to facilitate sludge hydrolysis and improve the stages of anaerobic digestion[4][9]. The effectiveness of these processes can be enhanced by employing a diverse array of microbes that work synergistically to break down the sludge floc structure and other organic substances [83].

2.3.1 Aerobic Pretreatment

Aerobic pretreatments (AD) can be implemented by treatment of sludge with air and aerobic or

facultative anaerobic microbes before AD [4]. Where, aerobic digestion consumes organic substance in waste and normally generates CO₂, H₂O and nitrate [53].

As part of the micro-aeration process, oxygen addition into system accelerates difficult organic substance hydrolysis through boosting hydrolytic actions of endogenous microbial population. Both aerobic and facultative anaerobic microbes, the presence of oxygen increases their hydrolytic activity. These microorganisms are significant biological resources which are utilized for SS pretreatment before AD. Micro-aeration pretreatment motivates exoenzymes excretion which slowly biodegrade substrates matter that otherwise stay

recalcitrant under anaerobic environments. Moreover, micro-aeration before AD in several studies improves AD hydrolysis step and also rises the overall methane yield [4].

Aeration can be achieved using open flasks, compressed air, air stones, or oxygen injection. Conversely, the autohydrolysis process involves experiments that use aerobic conditions in thermophilic reactors prior to anaerobic digestion in mesophilic reactors [9].

2.3.2 Anaerobic pretreatment

Anaerobic digestion (AD) is generally recognized as a single-stage biological process, involving a series of reaction steps and anaerobic activities occurring within the same bioreactor, without any physical separation. To intensify and improve AD efficiency, a focus has been placed on augmenting the hydrolysis stage [9].

As shown in Fig. 6 [53], anaerobic digestion includes four stages: hydrolysis, acidogenesis, acetogenesis, and methanogenesis. On other hands, chemical composition, substrate characterization, biodegradability, and operational parameters (pH, temperature, retention duration, loading rate, Carbon to Nitrogen (C/N) ratio, etc.) are significant factors in optimizing the anaerobic co-digestion process of biogas production technology [84].

Anaerobic processes is performed by substrates predigesting in mesophilic or thermophilic environments [4]. While thermophilic anaerobic digestion is superior to mesophilic anaerobic digestion for sludge disintegration, whereas thermophilic digestion has retention time of 10 to 12 days, conversely from 20 to 30 days for mesophilic digestion. So thermophilic process leads to higher pathogen inactivation and biogas yield than mesophilic anaerobic digestion. Hence, AD is changed to thermophilic anaerobic digestion in sludge handling units [83].

Therefore for anaerobic pretreatment, the most common technology is temperature phased anaerobic digestion (TPAD), that utilizes hydrolysis under thermophilic conditions (55 °C) or hyper

thermophilic (60–70 °C) conditions [6], followed by mesophilic digester (worked at about 35 °C). Typically the processes are worked in sequence, supplying raw material into the first thermophilic digester, then moving it to the second mesophilic digester. It is possible to separate the four steps of the bio-methanation process: polymeric raw material hydrolysis and subsequent acidogenesis happen primarily in the thermophilic digester, but acetogenesis and syntrophic methanogenesis happen in the mesophilic digester [1]. Using dual temperatures improves sewage sludge hydrolysis and acidogenesis in thermophilic conditions while promoting acetogenesis and methanogenesis in mesophilic conditions. This pretreatment approach is also called as two stage anaerobic digestion. TPAD process showed many benefits such as disintegration enhancement in floc and solid structure, production more biogas, requirements of thermal energy in low quality, as well as pathogens killing through thermophilic digestion [4].

2.3.3 Enzyme-assisted pretreatment

Enzymatic hydrolysis is considered a natural procedure driven by extracellular enzymes generated by sludge biomass, acting as effective catalysts for biochemical reactions leading to sludge solubilization. This stage of anaerobic digestion can be speeded by adding exogenous enzymes, which facilitate the decomposition and usage of organic matters for producing biogas. Within a few hours, these enzymes can change cell wall components into lower molecular weight compounds, making them easier to digest [1].

Addition of hydrolytic enzymes in pretreatment process leading to degradation of EPSs, sludge solubilization improvement, and rises production of biogas. On other hand, specificity, activity, quantity, temperature, enzyme stability and pH are the parameters which should be optimized and evaluated for successful enzymatic pretreatment [4], besides combination of suitable options in terms of pH, enzyme mixture kind, dosage, ratio, and ranges of temperature, is required [9].

Many researches have assessed industrial enzymes influences either added in isolation for example enzymes of proteases, lysozymes, and amylases or in the shape of enzyme pools comprising various

mixtures of proteases, lysozymes, lipases, amylases and cellulases on both primary and secondary sludge digestion, verifying well biodegradability and disintegration after the enzymatic hydrolysis [1].

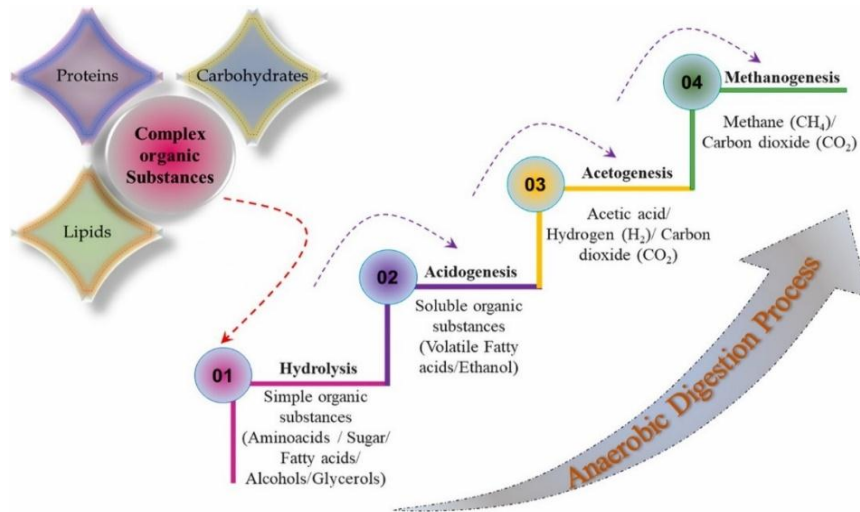


Fig. 6. Stages of anaerobic digestion to biogas generation

Table 5: Recent researches on biological pretreatment methods.

Method	Conditions	Results	Ref.
Aerobic pretreatment	Rice straw (RS) pretreated with cow manure (CM), sheep dung (SD), liquid consortium (LC), biogas slurry (BS), and straw-decomposing consortia (SC) at oxygen concentrations of 0, 6, 12 and 24 mL O ₂ /g VS.	largest cumulative methane production of 311.7 mL/g VS for pretreatment of RS with SC at optimal oxygen concentration of 12 mL O ₂ /g VS.	[85]
	T = 35 °C for 48 h with an aeration flow of 0.35 vvm, addition of fly ash or copper mining residues as trace metal supplementation.	201.6% increase in methane for fly ash, 185.8% for mining residues respect to control.	[86]
	Autohydrolysis pretreatment of secondary sludge: T = 55 °C, for 12–24 h with a limited oxygen.	Organic matter solubilization raised by 40%, methane production improved by 23% (12 h).	[87]
	Aerobic and aerobic/anoxic stabilization processes.	At the 30 th day of aerobic stabilization, removal of TOC, SS, and VSS = 55%, 22% and 28% respectively, while under aerobic/anoxic stabilization, removal of TOC SS, and VSS = 67%, 25% and 27% respectively.	[88]
Anaerobic pretreatment	TPAD-I reactor: thermophilic at	VS reduction = 77% for TPAD-1 and 72% for TPAD-II, methane production (45 °C , 3.55 ±	[89]

Enzyme-assisted pretreatment	45°C then mesophilic at 35 °C ; TPAD-II reactor: thermophilic at 55°C then mesophilic at 35 °C.	0.47 L CH ₄ L ⁻¹ day ⁻¹ ; 35 °C , 1.44 ± 0.12 L CH ₄ L ⁻¹ day ⁻¹) remained higher in TPAD-I than TPAD-II.	
	TPAD: SRTs of 14 - 20 days.	TPAD achieved (39 - 45%) higher methane, respect to single stage mesophilic digesters.	[90]
	TPAD systems: at 50–65 °C for HRT of 2 days for thermophilic pretreatment before 35 °C digestion a thermophilic–mesophilic TPAD against a mesophilic–mesophilic TPAD for primary sludge.	Thermophilic–mesophilic TPAD showed VS destruction of 54% with respect to 44% in mesophilic–mesophilic TPAD, with a 25% growth in methane yield.	[91]
	Two-step process for primary sludge: hyper thermophilic hydrolysis at 70 °C, HRT = 2 days then a thermophilic anaerobic digestion at 55 °C, HRT = 13 days, one-step (as reference process): at 55 °C, HRT =15 days.	The two-step process gave 12% higher in removal efficiency of organic suspended solids and well pathogen decrease impact than traditional one step digestion.	[92]
	Enzymes pretreatment of endogenous amylase and endogenous protease, also combined amylase/protease.	SCOD and VFAs raised by 78.2% and 129.6%, for endogenous amylase after 7 h respectively, while 23.1% enhancement in biogas yield compared to raw sludge for combined enzyme.	[93]
	Enzymatic solubilization (mixture of protease (5%) and lipase (5%)) and thermal solubilization (95 °C) associated in series.	Biogas production potential reached 291 ± 3 NmL/g VSS with increase in production of 71%.	[94]
	Studying cellulase enzyme, SRT and temperature effect on improvement of primary and rotating belt filter (PS, RBF) sludges fermentation.	Cellulose enzyme improved VFA productions by 86% and 36% for RBF and PS sludge, respectively.	[95]
	Fungal mash rich in hydrolytic enzymes was utilized in activated sludge pretreatment, food waste pretreatment and their combination before AD.	VS reduction = 19.1% for activated sludge, and 21.4% for food waste by fungal mash.	[96]

2.4 Combination pretreatment

The utilization of single pretreatment techniques are popular methods as standalone processes but the successive or simultaneous implementation for some various techniques has been examined [9]. Whereas most pretreatment methods utilize much energy or have very installation, working, and O&M costs. Therefore, the combination of various chemical,

physical and mechanical pretreatments is considered a well option to augment sludge solubilization, methane generation, and volatile solids decrease [1]. Hence, several studies have been employed to assess these combined pretreatments which commonly divided into thermo-chemical, mechanical-thermal and mechanical-chemical combinations [9].

Thermo-chemical method contains thermal distraction with the simultaneous adding of acid

alkaline or oxidative matters. Chemical agents are supplemented to the pretreatment separately or in combination for example NaOH, KOH, HCl, O₃, and H₂O₂ [6]. As a result, Thermochemical pretreatment achieved higher volatile solids solubilization than singular thermal and chemical treatments [1].

Mechanical-chemical combination showed considerable results of effectiveness such as ultrasonication combined with ozonation, Fenton's oxidation, and alkaline pretreatment or with microwaves. Hence, the alkaline addition has a pivotal role for COD solubilization. So, numerous studies focused on combined mechanical with alkaline pretreatment. While, mechanical-thermal

combined pretreatment has advantage of the shear forces produced by mechanical techniques to disrupt the cell walls and disintegrate structure of sludge, as well as heating which softens the sludge components and elevates the solubilization before AD [9].

Table 6: Recent researches of combination pretreatment methods.

Method	Conditions	Results	Ref.
Thermo-chemical combination	Low temperature-alkaline: T = 60 °C, 60 min, low dose of 16 g NaOH/kg TS, WAS.	DD = 28.7 %, methane raised from 210.4 ± 10.9 (raw) to 248.8 ± 13.1 and 267.1 ± 14.3 mL/g VS for HRT of 15 and 21.5 d respectively.	[97]
	Thermal-alkaline: T = 90 °C, pH = 11 for 10 h, WAS.	Degree of solubilization was 42.29%,	[98]
	Alkaline-thermal pretreatment at pH = 12, T = 75 °C.	9.6% TS solubilization, 17.2% VS solubilization.	[99]
	Alkaline-thermal: pH = 12 (NaOH) then heated at 134 °C, 3.4 bar, 30 min	DD% = 37.07 ± 0.19 %.	[67]
	WAS pretreatment by thermo-alkaline: 105 °C, pH 10.0 for 2 h.	COD solubilization = 26 % than COD solubilization (10.5 %) of raw sludge at pH 7.	[100]
	Thermal-alkaline: pH = 12 for 1 h then thermally treated 121 °C for 1 h.	134% increase in SCOD, 38% of VSS content were solubilized.	[101]
Mechanical-chemical combination	HC (orifice: 27 holes, 1 mm) combined with alkaline (pH 11).	Maximum disintegration degree = 48.4%.	[25]
	Combined MW (600W, 85 °C, 2 min) with alkaline (pH 12, 30 min), WAS.	Sludge solubilization improved to 46%.	[102]
	Electrolysis (5 V) with alkaline (pH = 9.2 by NaOH) pretreatment for AD promoting (42 days).	DD _{SCOD} jumped from 0.1 ± 0.2% (electrolysis alone) to 38.7 ± 0.3% at pH = 12.2, 20.3% methane production improvement.	[103]
	HPH (P = 0 - 12,000 psi), chemical dose of 9–36 mg NaOH/g TS.	SCOD/TCOD = 27% for P=12,000 psi and dose of 36.0 mg NaOH/g TS.	[104]
	Electrochemical (EC) combined with hydrogen peroxide, WAS.	DD _{COD} reached 11.80 %.	[105]
	H ₂ O ₂ = 0.5 mmol/L, pH = 11, sludge concentration = 17 g/L, 15 min US	SCOD = 3662.78 ± 239.21 mg/L, with DD of 28.61 ± 2.14%, and ΔMLSS = 3.31 ± 0.06 g/L.	[106]

	Photo-Fenton: 725 g H ₂ O ₂ /kg TS, H ₂ O ₂ /Fe ⁺² = 80, 40 min irradiation.	MLVSS removal, EPS and SCOD reached 25%, 500 mg/L and 12,000 mg/L, respectively.	[107]
	Electrochemical-Fenton process: 20 min, H ₂ O ₂ of 25 mg/g DS, Fe ⁺² = 15 mg/gDS, voltage = 11 V	Reduction in specific resistance to filterability and time to filter parameters was 93.8% and 75.9%, respectively.	[108]
Mechanical-thermal Combination	Combined US- thermal pretreatment 5,000 kJ/kg TS, 20 kHz, 30 sec; then followed by thermal at 65°C.	SCOD increased from 760 to 10,200 mg/L, proteins increased from 115 to 2,900 mg/L, 20% increase in biogas production.	[109]
	Thermal combined ultrasonic: 100°C 1 h then sonication of 40 kHz for 1 h.	SCOD represents 32 times of the control, Biogas yield raised by 19%, CST decreased by 85%, VS removal rate enhanced above 50%.	[110]

Table 7: Advantages and disadvantages of various pretreatment techniques in short summary [1][2][3][4][9][6][83] [11][19][20][53][111].

Method	Advantages	Disadvantages
1. Physical pretreatment: Cavitation	1- Low O&M cost, simple construction, and easy operation especially orifice and venturi 2- Sludge settleability improvement 3- Particle size reduction 4- Rise in surface area 5- Sludge solubilization	1- Clogging and erosion trouble for orifice and venturi 2- High O&M cost especially for rotating type.
Ultrasonication	1- Compact design and easy maintenance 2- Quick process and real-scale applications 3- No odor generation or clogging problems 4- Reduction in particle size and pathogens 5- No recalcitrant compounds generation 6- Well dewaterability 7- Well methane generation 8- Effective cellular wastes degradation	1- High capital and working costs and extra energy usage 2- Restricted applicability 3- Inappropriate for lignocellulosic biomass 4- High-tech process
Microwaves	1- Quick and regular heating process 2- Easy to control and no odour production 3- Cost-effective (energy, space, and time) 4- Hazardous emissions and pathogens reduce 5- Sludge dewaterability improvement 6- Very effective on biogas production	1- Extreme energy requirements 2- Scale up challenges 3- Lack of data availability and unavailability of full-scale experience 4- Limited to microbial cell-based substrates
Electric decomposition	1- High-tech and compact process 2- Small process time 3- Rise in soluble components 4- Raised biogas and methane production 5- TCOD and VS removal 6- Medium effect on foaming during digestion	1- High energy need 2- Working side-effects like electrode corrosion 3- Difficult operation and maintenance 4- Shortage of research and limited application in the field
Thermal pretreatment	1- Relatively simple method 2- Reduce in sludge volume 3- Improved dewaterability and settleability 4- Sludge sanitation because of pathogen removal 5- Reduce in sludge viscosity. 6- Better sludge solubilization and Well impact for biogas	1- High O&M costs and odour production 2- Heat exchangers fouling 3- High energy requirement and capital cost 4- Possible generation of complex substrates that are hard to biodegrade 5- Rise in soluble inert fraction, 6- Raised ammonia inhibition in the basic digester.

2. Chemical

pretreatment: Alkaline/acidic pretreatment	1- Simple and easy operation 2- Low-cost pretreatment 3- Sludge pathogens reduce 4- Increased methane yield 5- Acidic pretreatment is effective for lignocellulosic biomass 6- Alkaline is suitable for lignin breakdown 7- Alkaline enhances sludge suitability and COD solubilization	1- High quantity and cost of chemicals 2- Toxicity of some alkali like Na^+ 3- Bad odour formation 4- Especial materials for reactor construction 5- Risk of recalcitrant compounds creation 6- Fouling and corrosion of the equipment 7- Makes inhibitor effects on AD 8- Sludge needs subsequent neutralization
Ozonation	1- Flexible operation and small reaction time 2- Reduce in sludge volume 3- No addition of chemicals 4- More sludge solubilization 5- Very effective on biogas generation 6- Better dewaterability and settleability 7- Sludge pathogens reduce 8- Ozone produced on-site	1- High consumption of energy essential for ozone generation 2- Partial or full mineralization of liberated cellular matters, influencing on maximum methane yield 3- Raised polymer demand for dewatering 4- Creates non-biodegradable components 5- Limited application
Fenton oxidation	1- Low energy requirement. 2- Plain device and easy operation. 3- Sludge solubilization enhancement 4- Increased biodegradability 5- Non-toxic nature 6- Increased methane yield 7- Good effective on biogas yield	1- High chemicals cost 2- Highly corrosive (due to very low pH). 3- Chemical pollution, 4- Scavenging hydroxyl radicals danger 5- Sludge neutralization before digestion 6- Generation of Fenton sludge
3. Biological pretreatment: Aerobic pretreatment	1- Volatile solid reduction 2- Accelerated hydrolytic activities 3- Improvement in sludge solubilization 4- Increase in methane yield	1- Required equipment to ensure quantity of the air provided 2- More installation of aeration device may rise costs 3- Slow procedure 4- Needs suitable conditions for microbial activities
Anaerobic pretreatment	1- Improved floc and solid structure disintegration 2- Volatile solid reduction 3- No additional chemicals 4- Kills pathogenic microbes 5- Low thermal energy 6- Increase in methane yield	1- Needs factor optimization 2- Supplementary equipment installation for energy balance may rise costs 3- Slow process 4- Biokinetic factors for microbial communities should be assessed.
Enzyme- assisted pretreatment	1- Low energy need. 2- Degradation of polymeric substances 3- Sludge solubilization enhancement 4- Well effective on biogas production	1- Various factors should be optimized before addition enzymes 2- Enzyme specificity with the substrate should be measured 3- Further development requirement 4- Extreme enzyme price and slow process 5- Requires adding enzyme continuously 6- Unfeasible at a full-scale
4. Combination pretreatment	1- High admiration to good efficiencies in terms of sludge hydrolysis, organic components solubilization, biogas, and methane production than these techniques of single methods	1- High operation and maintenance cost 2- High energy consumption

3 Summary and future research

This review is concentrated on different sludge pretreatment methods and their effectiveness on sludge disintegration and improving its properties, but most of these methods is conducted in lab scale with small models so the future approaches should focus on the full scale application in treatment plants using the massive amounts of sludge to produce more biogas in order to recover the consumed energy to minimize the total cost. Besides, achieving the difference in the results between the lab and field application. Also the future research must be more effective in new approaches such as studying recent technologies like usage the nano technology materials for sludge pretreatment or combined it with other physical methods like hydrodynamic cavitation to increase its efficiency. Investigating the long-term impacts of chemical residues from pretreatment on sludge quality and their impacts on anaerobic digestion efficiency. Optimization the consumed energy specially through thermal and mechanical methods through optimization the effective parameters on the usage of energy. Furthermore, more studies should focus on the cavitation process to solve the problems of clogging or erosion in some cavitation devices.

4 Conclusion

Generally, several pretreatment methods can pretreat sludge and other wastes before AD process such as physical, chemical, biological and combination between these methods, leading to an improvement in sludge dewaterability, disintegration degree increase and biogas production enhancement through AD process. Where the physical methods showed more enhancement in sludge disintegration than other approaches, since both cavitation and thermal pretreatment demonstrated an effective improvement in disintegration degree. Alkaline pretreatment (specially at pH of 12 by NaOH) has a great disintegration effects among other chemical methods. So, the maximum disintegration values were achieved through the combined physical-chemical methods, where the thermal-alkaline pretreatment at temperatures above 90°C showed high effectiveness in sludge solubilization, while mechanical-chemical combinations offered moderate cost-effectiveness than mechanical-thermal methods. Therefore, Egypt should utilized the sludge produced from many treatment plants for AD process to generate more biogas production, which is regarded as fuel source to face the challenges and problems in field of energy and electricity to cover the increased consumption.

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