Ultrasonics Sonochemistry xxx (2010) xxx–xxx

Contents lists available at ScienceDirect

Ultrasonics Sonochemistry

journal homepage: www.elsevier.com/locate/ultsonch

Review

Ultrasonic pretreatment of sludge: A review

Sridhar Pilli [a], Puspendu Bhunia [b], Song Yan [a], R.J. LeBlanc [c], R.D. Tyagi [a,*], R.Y. Surampalli [d]

[a] INRS Eau, Terre, Environnement, 490, rue de la Couronne, Québec, Canada G1K 9A9
[b] Dept. of Civil Engineering, Indian Institute of Technology, Bhubaneswar 751 013, India
[c] GMSC, 355 Hillsborough Road, Riverview, NB, Canada E1B 1S5
[d] US Environmental Protection Agency, P.O. Box 17-2141, Kansas City, KS 66117, USA

ARTICLE INFO

Article history:
Received 15 June 2009
Received in revised form 24 December 2009
Accepted 1 February 2010
Available online xxxx

Keywords:
Anaerobic digestion
Biological wastewater treatment
Cavitation
Waste activated sludge
Ultrasonication

ABSTRACT

Ultrasonication is an emerging and very effective mechanical pretreatment method to enhance the biodegradability of the sludge, and it would be very useful to all wastewater treatment plants in treating and disposing sewage sludge. Ultrasonication enhances the sludge digestibility by disrupting the physical, chemical and biological properties of the sludge. The degree of disintegration depends on the sonication parameters and also on sludge characteristics, therefore the evaluation of the optimum parameters varies with the type of sonicater and sludge to be treated. The full-scale installations of ultrasonication have demonstrated that there is 50% increase in the biogas generation, and in addition evaluation of energy balance showed that the average ratio of the net energy gain to electric consumed by the ultrasound device is 2.5. This review article summarizes the benefits of ultrasonication of sludge, the effect of sonication parameters, impact of sludge characteristics on sludge disintegration, and thereby the increase in biogas production in anaerobic digester. Due to uncertainty in the unit representation by many researchers and nonavailability of the data, comparison of these results is complicated. Comparison of ultrasonication with other pretreatment options is necessary to evaluate the best economical and environmental pretreatment technology for sludge treatment and disposal. The optimum parameters for the ultrasonication vary with sludge characteristics.

1. Introduction

The rapid growth of industrialization and urbanization in the 21st century has resulted in production of unmanageable quantity of sludge from wastewater treatment plants (WWTPs). The sludge management is the major issue of wastewater treatment plant, as it costs 60% of the total plant capital cost [1] and the laws for sludge disposal are becoming increasingly stringent. With increase in global warming and climate change, the greenhouse gases (GHGs) emissions from the waste sector are of increasing concern. In Canada, GHGs from the waste sector have increased by 15% from 1990 to 2006 [2]. The existing WWTPs in Canada are producing 670,000 Mg/y of dry sludge [3], and the production rate is expected to increase further in the future. Incineration, ocean discharge, land application and composting are the common sludge disposal methods used over the years. These common sludge disposal methods are no longer reliable due to the economical constraints and the negative impacts on environment. Due to environmental and economical constraints, there is a need for affordable and sustainable technologies for sludge treatment and disposal. With extensive research on sludge treatment, many researchers have proposed anaerobic digestion (AD) of sludge as the efficient and sustainable technology for sludge treatment. The benefits associated with AD technology are huge, which include mass reduction, odour removal, pathogen reduction, less energy use, and more significantly, the energy recovery in the form of methane.

The AD of sludge is a complex process that converts degradable organic compounds to methane (CH_4) and carbon dioxide (CO_2) in the absence of elemental oxygen with a series of microbiological process. The conversion pathway of the substrate to biogas (mainly CH_4 and CO_2) occurs in four stages, namely, hydrolysis, acidogenesis, acetogenesis and methanogenesis by three different groups of bacteria. The first group involves hydrolytic and acidogenic bacteria, which hydrolyze the complex substrates (carbohydrates, lipids, proteins, etc.) to dissolved monomers (sugars, fatty acids, amino acids, etc.) and further to CO_2, H_2, organic acids and alcohols. The second metabolic group of bacteria is hydrogen producing acetogens that convert the simple monomers and fatty acids to acetate, H_2, and CO_2. The third group is methanogenic bacteria that utilize the H_2, CO_2 and acetate to produce CH_4 and CO_2. This complete microbial digestion process of the substrate to CH_4 and CO_2 is a slow process and requires high retention time. In particular, intracellular biopolymers solubilisation and conversion to the lower molecular weight compounds of solid degradable organics such as sludge through hydrolysis is a rate limiting step [4,5]. The

---

* Corresponding author. Tel.: +1 418 654 2617; fax: +1 418 654 2600.
E-mail address: tyagi@ete.inrs.ca (R.D. Tyagi).

1350-4177/$ - see front matter © 2010 Elsevier B.V. All rights reserved.
doi:10.1016/j.ultsonch.2010.02.014

Please cite this article in press as: S. Pilli et al., Ultrasonic pretreatment of sludge: A review, Ultrason. Sonochem. (2010), doi:10.1016/j.ultsonch.2010.02.014

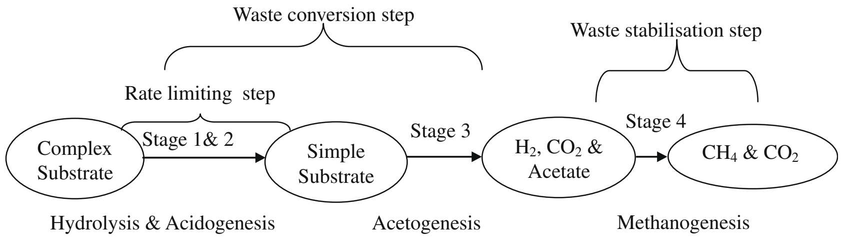

Fig. 1. Different stages of anaerobic digestion.

conventional anaerobic digestion process with four stages is presented in Fig. 1.

The lower microbial conversion rates during conventional AD process results in high hydraulic retention time in the digester and larger digester volume, which are the prime drawbacks of the conventional AD technology. The nonavailability of the readily biodegradable soluble and organic matters and lower digestion rate constant (i.e., first order digestion rate constant 0.15 day^{-1} for sludge) [6] necessitates the pretreatment of sludge. Pretreatment of sludge is required to rupture the cell wall and to facilitate the release of intracellular matter into the aqueous phase to increase biodegradability and to enhance the anaerobic digestion with lower retention time and with higher biogas production [7–9]. With the advancements in various sludge pretreatment techniques like thermal, chemical, mechanical, biological and physical and several combinations such as physicochemical, biological–physicochemical, mechanical–chemical and thermal–chemical, biodegradability of sludge can be enhanced by several orders; however, economical constraints of these technologies have limited their scale-up and lab-to-field implementation.

Extensive research has been carried throughout the world to establish the best economically feasible pretreatment technology to enhance the digestibility of sludge. The ultrasonication is an emerging and promising mechanical disruption technique for sludge disintegration due to several inherent merits like efficient sludge disintegration (>95%, [10]), improvement in biodegradability [9], improved biosolids quality [9], increase in methane percentage in biogas [9,11], no chemical addition [12], less retention time [13], sludge reduction [14] and energy recovery (1 kW) of ultrasound energy generates 7 kW of electrical energy including losses [11]. The order of pretreatment efficiency for enhancement of methane generation is: ultrasonic lysis (20 W, 9 kHz, 30 min) > thermal pretreatment by autoclave (120 °C, 30 min) > thermal pretreatment with hot water (60 °C, 30 min) > freezing (−10 °C, 15 h) [15]. This paper presents an extensive review of the ultrasonic pretreatment of sludge to enhance the AD, and to compare the results of the full-scale and lab-scale implementation.

2. Ultrasonication

In early days of sonar, the sound waves are used for anti-submarine warfare, results in the killing of fish by the sound waves, has given the birth of ultrasound method of destroying or inactivating biological cells. Hughes and Nyborg [16] and Alliger [17] have studied the mechanism of ultrasound interaction with microbial cells and observed that the brief exposure to ultrasound can cause thinning of cell wall attributed to releasing of the cytoplasm membrane from the cell wall. The ultrasound is cyclic sound pressure (compression and expansion) with a frequency greater than 20 kHz. The ultrasound range diagram with various applications at different frequency is shown in Fig. 2.

Depending on the frequency, it is divided into three regions: power ultrasound (20–100 kHz), high frequency ultrasound (100 kHz–1 MHz), and diagnostic ultrasound (1–500 MHz). Ultrasound application in medicine started during the Second World War as ultrasound massage to substitute for hands of the masseur in patients who has suffered from fractures [18]. With advancements in technology and sophistication, ultrasound (>20 kHz) is used in several fields. Ultrasound ranging from 20 kHz to 100 kHz is used in chemically important systems in which chemical and physical changes are desired [19,20]. The ultrasound ranging from 1 MHz to 10 MHz is used in different fields, like animal navigation and communication, detection of cracks or flaws in solids and under water echo location, fetal scanning, detection of pelvic abnormalities, treating benign and malignant tumors, etc. [19,21,22]. Biological cell disruption for the recovery of intracellular materials is achieved by ultrasonication [22–24], and further full-scale application for municipal sludge disintegration was evaluated by Hogan et al. [25]. The acoustic waves between 20 Hz and 20 kHz are the audible range, while hearing varies with the individual and the age. The acoustic waves less than 20 Hz down to 0.001 Hz is used in seismology [26], medical application [27] [ballistocardiography and seismocardiography to study the mechanics of the heart], and also in charting rock and petroleum formations below the earth [26].

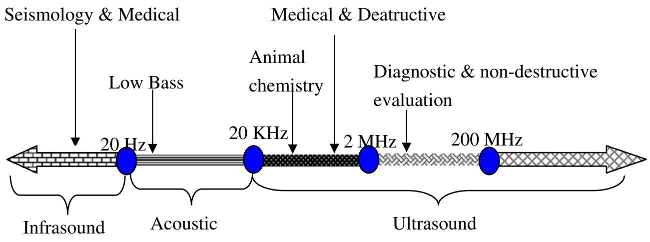

Fig. 2. Diagram of ultrasound range.

2.1. Cavitation phenomena induced by ultrasound

The basic goal of ultrasound technique is to spiflicate bacterial cell walls and to facilitate intracellular matter available for subsequent degradation to CH_4 and CO_2 in AD. When the ultrasound wave propagates in sludge medium, it generates compressions and rarefactions, the compression cycles exert a positive pressure on the liquid by pushing the molecules together and the rarefaction cycle exerts a negative pressure by pulling the molecules from one another. Because of this excessively large negative pressure, microbubbles (cavitation bubbles) are formed in the rarefaction regions. These microbubbles grow in successive cycles and reaches to an unstable diameter that collapse violently producing shock waves (temperature of around 5000 °C and pressure of 500 atmospheres at a lifetime of few microseconds) [14,28,29–34]. This process by which the bubbles form, grow and undergo violent collapse is known as cavitation. Representation of development and collapse of the cavitation bubble is shown in Fig. 3.

Please cite this article in press as: S. Pilli et al., Ultrasonic pretreatment of sludge: A review, Ultrason. Sonochem. (2010), doi:10.1016/j.ultsonch.2010.02.014

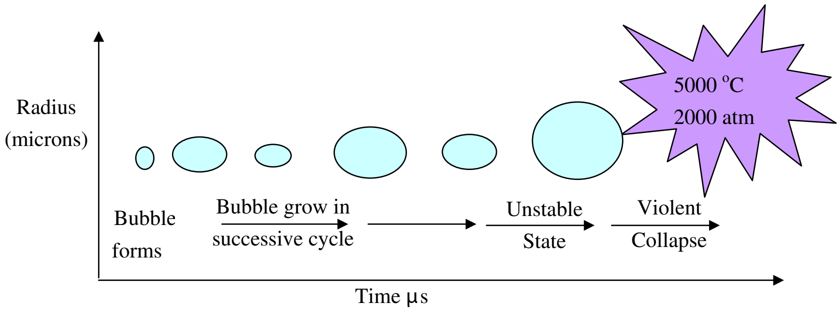

Fig. 3. Development and collapse of the cavitation bubble.

2.2. Factors influencing the cavitation phenomena

The sludge disintegration efficiency is essentially based on the cavitation phenomena and the factors influencing the cavitation that influence the efficiency of disintegration are shown in Table 1.

2.3. Ultrasound generation and disintegration mechanism

Ultrasound is generated by two techniques magnetostrictive and piezoelectric. In magnetostrictive technique, the electrical energy is converted to mechanical energy (or vibration) with a magnetic coil attached to vibrating piece like nickel and Terfenol-D [44]. In the piezoelectric technique, the electrical energy is converted to high frequency electric energy with piezoelectric crystals (rely to material strain) attached to the vibrating piece (sonotrode, probe or horn). The transducer (converter) converts the electrical or mechanical energy to sound waves and the booster is a mechanical amplifier that increases the vibration (amplitude) generated by transducer [9,45]. The horn delivers the ultrasound waves into the liquid. Therefore transducer, booster and horn are the major components in ultrasound equipment. Ultrasonication of sludge with temperature control and sonication components is shown in Fig. 4.

Holding or affixing the transducer, booster and horn together is called as stack assembly and the stack is clamped at nodal points as shown in Fig. 4. The two most common places to clamp or to hold the total system is at the transducer or at the booster nodal ring. Similar to the booster, the horn which delivers the motion to the sludge often amplifies the motion even further. In addition, the horn is usually half a wavelength long, but full wavelength designs are also common depending upon the application. The intensity of sonication can be controlled by altering the power input and is based on the probe design. This is a very important parameter in ultrasonication and will determine the magnitude of the gain or mechanical amplification of the vibration. The conversion efficiency of sound energy to thermal energy can be calculated as shown in Eqs. (1)–(3) [46].

Table 1
Factors influencing the cavitation phenomena.

No.	Factors	Influence on cavitation phenomena
1	Gas and particulate matter	Presence of gas/air in the liquid will lower the cavitational threshold and reduces the intensity of the shock wave released, as much of the shock wave will be utilized to collapse the gas bubbles. Particulate matters, especially like trapped vapour gas nuclei in their crevices and recesses, will reduce the cavitation effect [18]
2	External applied pressure	Increasing the external pressure raises the rarefaction pressure, which increases the cavitation collapse intensity [35,36]
3	Solvent viscosity	If the natural cohesive forces acting in the liquid are lower, then they will suppress [37] the negative pressure in the expansion or rarefaction cycle. Therefore to increase the cavitation threshold the natural cohesive forces need to be increased by increasing the viscosity of liquid
4	Solvent surface tension	The addition of surfactant to an aqueous solution certainly facilitates the cavitation. Increase in solvent viscosity and surface tension, reduces the rate of microbubble formation but increases the intensity of bubble collapse. With addition of surfactants will reduces the solvent surface tension and facilities bubble nucleation (i.e., fewer microbubbles are formed) [35,38]
5	Solvent vapour pressure	If the vapour pressure of the liquid is low, then it is difficult to induce cavitation in the liquid. Because, low vapour will enter into the bubble and results in low cavitation [37]
6	Applied frequency	The rarefaction phase is shortened by increasing the frequency of irradiation, but to maintain an equivalent amount of cavitational energy into the system the power should be increased. That is at higher frequency more power is required to maintain same cavitational effect [35,39,40]
7	Temperature	The cavitation threshold increases with decrease in temperature of bulk solution. With increase in temperature, the solvent reaches the solvent boiling point and produces larger number of cavitation bubbles concurrently, which acts as barrier to sound transmission and nullify the effectivity of ultrasound energy [38]
8	Sonication density	Increase in sonication density increases the sonication effects on the sludge as given by the equation, $P_A = \sqrt{2}I\rho C$, [41]; where P_A = acoustic pressure, I = intensity, ρ = density, C = velocity of sound in the medium
9	Acoustic intensity	Increasing the sonication intensity increases the sonication effects, and it is directly proportional to the square root of the amplitude (P_A) of the acoustic wave divided by the density of the liquid (ρ) and the speed of sound in the liquid (c). $I = \frac{P_A^2}{2\rho c}$ [36,42]
10	Types of ultrasound cavitation	The collapse of the cavitation bubbles produces high velocity waves and temperature, causing inter-particle collision and the rupture of cell wall. Depending on bubble types, the ultrasound cavitation is classified as transient or stable (non-inertial cavitation). Transient is believed to occur at 10 W/cm^2 and the later at 1–3 W/cm^2 [37]; the stable bubbles bound to have significant long term effect. The transient and stable bubble growth is explained by bubble growth time by Abramov [43]; $\tau_g = 0.75T + (i - 1)T$; $T = 1/f$, where τ_g is the bubble growth time, 'f' is the ultrasound frequency, 'T' is the period of ultrasound wave and 'i' is number of acoustic cycles the bubble experienced
11	Attenuation	The intensity of the ultrasound is attenuated as it progress through the medium. The attenuation is inversely proportional to the frequency of the ultrasound (i.e., energy is dissipated in form of heat which is not considered in the bulk medium). High power and high frequency is required to have the same intensity at the lower depth for a given sample
12	Field type	The standing wave field is pronounced with more acoustic cavitation than a progressive field [37]

Please cite this article in press as: S. Pilli et al., Ultrasonic pretreatment of sludge: A review, Ultrason. Sonochem. (2010), doi:10.1016/j.ultsonch.2010.02.014

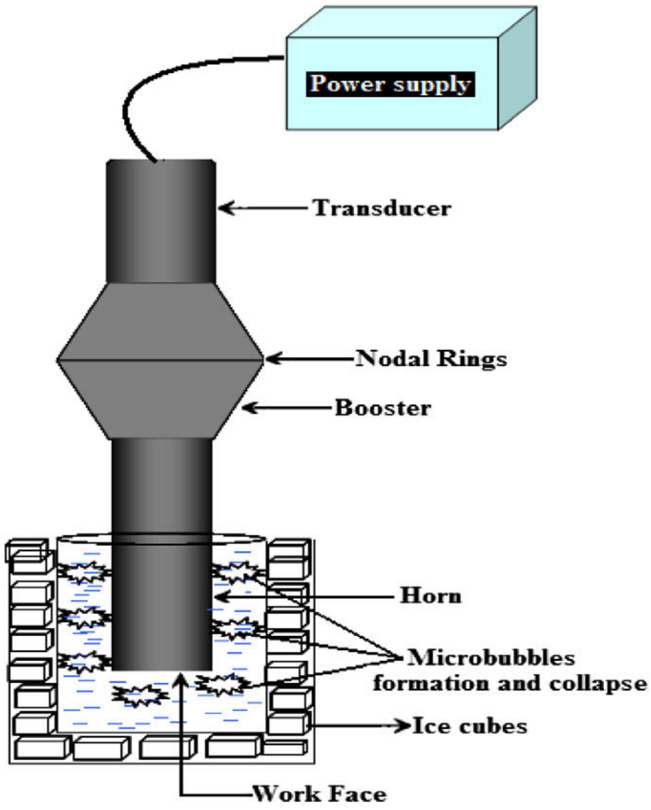

Fig. 4. Ultrasonication of sludge (temperature controlled).

$$Q_w = Cm(T - T_0) \tag{1}$$

$$Q_u = Pt \tag{2}$$

$$\eta(\%) = \frac{Q_w}{Q_u} * 100 \tag{3}$$

T_0 = temperature of fresh sludge; T = temperature after ultrasonication; Q_w = total energy; m = mass of water; C = specific heat of water (4.2 kJ/kg C); Q_u = actual energy produced; P = ultrasound power; t = ultrasound duration; η = efficiency.

2.3.1. Sludge disintegration

The applied power/energy supplied for sludge disintegration is expressed in many ways, (a) specific energy input, (b) ultrasonic dose, (c) ultrasonic density and (d) ultrasonic intensity and the expressions are given in Table 2.

The expected disintegration mechanisms during ultrasonic disintegration of sludge are as follows [9,33,49]:

(a) Hydro-mechanical shear forces
(b) Oxidising effect of $^{\cdot}$OH, $^{\cdot}$H, $^{\cdot}$N, and $^{\cdot}$O produced under the ultrasound radiation
(c) Thermal decomposition of volatile hydrophobic substances in the sludge
(d) Increase of temperature during ultrasonic activated sludge disintegration

Table 2
Expressions for sludge disintegration.

No.	Parameter	Expression	Unit	Reference
1	Specific energy input	$E_s = \frac{P*t}{V*T*S}$	kJ/kg TS or kW s/kg TS	[46]
2	Ultrasound dose	$UD_o = \frac{P*t}{V}$	J/L	[33]
3	Ultrasound density	$UD = \frac{P}{V}$	W/L	[33]
4	Ultrasound intensity	$UI = \frac{P}{A}$	W/cm^2	[48]

E_s: specific energy in kW s/kg TS (kJ/kg TS); P: power input (kW); T: sonication time (s); V: volume of sludge (L); TS: total solids concentration (kg/L); A: surface area of the probe in cm^2.

The induce cavitation that occurs during ultrasonication results in sudden and violent collapse of huge number of microbubbles, which generates powerful hydro-mechanical shear forces in the bulk liquid surrounding the bubbles [9,34,49–51]. The high temperature produced during the bubble collapse (implosion) decomposes water (H_2O) into extremely reactive hydrogen atoms (H^+), and hydroxyl radicals ($^{\cdot}$OH) and in the cooling phase these radicals will recombine to form hydrogen peroxide and molecular hydrogen [9,33,49,52–56]. The oxidising effect of $^{\cdot}$H, $^{\cdot}$N and $^{\cdot}$O is less than $^{\cdot}$OH [49,53,54,57]; therefore the effect of $^{\cdot}$H, $^{\cdot}$N and $^{\cdot}$O is neglected during the ultrasonication. The effect of the volatile hydrophobic substances is neglected as their quantity is very low in the sludge [49]. Considering the temperature effect on solubilisation, at higher temperature the sludge solubilisation is very low and needs longer time to attain the solubilisation of sludge (temperature raised to 80 °C in 1 h produces low solubilisation and a low degree of disintegration in the sludge, where the sonication is a short period of time). Therefore, sludge disintegration is expected to occur in two ways, mainly by hydro-mechanical shear forces and the oxidising effect of $^{\cdot}$OH.

Wang et al. [49] have evaluated the effect of $^{\cdot}$OH and hydro-mechanical shear forces on sludge disintegration. The effect of hydroxide radical was evaluated by addition of the $NaHCO_3$ to the sludge prior to sonication. The oxidation effect of hydroxide ion on sludge solubilisation was slightly higher by the addition of $NaHCO_3$, but the slight enhancement was due to the increase in the pH of the sludge. This shows that the oxidation effect of the hydroxide radical on sludge solubilisation is negligible. Therefore, the disintegration of the sludge occurs mainly by hydro-mechanical shear forces produced by cavitation bubbles. The sludge disintegration by hydro-mechanical shear forces and hydroxide radical follows a first order reaction. The total reaction constant "u" is calculated as follows (Eq. (4)) [49].

$$u = u_{\cdot\mathrm{OH}} + u_{\mathrm{HSF}} \tag{4}$$

where $u_{\cdot\mathrm{OH}}$ is the reaction constant under oxidising effect; u_{HSF} is the reaction constant under effect of hydro-mechanical shear forces. Neglecting the effect of hydroxide radical oxidation effect, then the reaction constant is $u = u_{\mathrm{HSF}}$. The reaction rate constant of hydroxide radical increases with increase in ultrasonic density; for example, the contribution of the oxidising effect of hydroxide radical increased from 19.15% to 25.86%, with increase in ultrasonication density from 0.384 to 0.72 W/mL [49].

2.4. Evaluation of ultrasound disintegration

The ultrasound de-agglomerates the biological flocs and disrupts the large organic particles into smaller size particles. The shear force produced by high pressure wave breaks down bacterial cell wall and releases the intracellular substances into aqueous phase. This changes the physical, chemical and biological properties of sludge during pretreatment by ultrasonication. Therefore, the degree of sludge disintegration is to be evaluated based on the changes in physical (particle size distribution, turbidity, settleability, mass composition and microscopic examination), chemical (increase in SCOD, protein concentration, polysaccharide content of the supernatant, nitrate nitrogen and release of NH_3)

Please cite this article in press as: S. Pilli et al., Ultrasonic pretreatment of sludge: A review, Ultrason. Sonochem. (2010), doi:10.1016/j.ultsonch.2010.02.014

and biological (heterotrophic count and specific oxygen uptake rate) properties.

2.4.1. Physical changes

The physical parameters of the sludge have a significant effect on AD, so the evaluation of the physical parameters after sonication is essential for operation of AD. Further, physical evaluation is used as qualitative measurement of sludge disintegration. Particle size analysis, sludge settleability, mass composition, microscopic image, turbidity, and sludge dewaterability are some of the techniques used to judge the degree of ultrasonic disintegration. Particle size is analysed by various techniques depending upon size of particles. The different techniques are sieves, sedimentation, electrozone sensing, microscopy, laser diffraction [58,59]. Ultrasonication disintegrates sludge particles to a very smaller size, and laser diffraction is usually used for particle size analysis. The turbidity of sludge changes with increase in sonication parameters (ultrasound density, ultrasound intensity, and sonication time) and it is measured by using a turbidity meter, with NTU units. The sludge dewaterability is measured based on the capillary suction time (CST) and specific resistance of filtration (SRF) [13,60–65].

2.4.1.1. Particle size. The particle solubilisation rate is governed by the size of particles in the waste, and methane production in the mature digester is proportional to the net rate of particle size solubilisation [5,51,66]. Ultrasound pretreatment is very effective in reducing the particle size of sludge and the efficiency of size reduction is dependent upon the sonication duration [13,67–70], ultrasonication density [12,41,61,64,71–75], sonication power [41,59,62,73], sludge volume and sludge characteristics [12,47,66,69,72,73]. With increase in sonication time the particle size reduces gradually; for example, the particle size reduces from 165 μm to 135 μm and 85 μm with a sonication time of 0.49 min and 1.6 min, respectively [13]. Similarly, Biggs and Lant [68] observed a particle size reduction of 125–10 mm after 5 min sonication (50 W, 100 ml). Gonze et al. [69] observed a similar reduction trend initially but beyond a sonication of 10 min the particle size has increased gradually with further increase in sonication time. At 0.5 min @ 3.9 kJ/L, 1 min @ 7.8 kJ/L, 3 min @ 23.4 kJ/L and 6 min @ 46.8 kJ/L, the particle size was reduced from 66.9 μm to 55.1 μm, 42.6 μm, 24.2 μm and 18.1 μm, respectively, but beyond a sonication time of 10 min the particle was increased gradually to 19.5 μm, 20.6 μm and 31.9 μm at 10 min @78 kJ/L, 20 min @ 156 kJ/L and 60 @ 468 kJ/L min, respectively.

The increase in the particle size at higher sonication time is due to re-flocculation of the particles. Initially the flocs are reduced but increase in sonication time further causes more release of intracellular polymers due to cell lysis that are favourable for re-flocculation [64,67,69,76,77]. The biopolymers released are thought to be the glue that holds bioflocs together and they form functional groups such as hydroxyl and negatively charged carboxy groups [78]. The mean particle size reduction also increases with increase in sonication densities. At densities of 0.52 W/mL, the mean particle is reduced from 51 μm to 15 μm and 51 μm to 19 μm at 0.33 W/mL, respectively [72]. Low power level has no effect on floc size reduction by sonication. With increase in power level, the floc size reduction increases with increase in ultrasonication density and sonication time. For example, the floc size reduces from 94 μm to less than 3 μm with a sonication density of 0.22 W/mL and 0.44 W/mL, respectively [61]. At constant power level of 0.33 W/mL, with increase in sonication time reduces the floc size to 22 μm and 10 μm with a sonication time of 20 min and 40 min, respectively [61]. At constant sonication time of 60 min, the particle size reduction was less than 3 μm at 0.33 W/mL and 0.44 W/mL. Microscopic examination reveals that structural integrity of floc will be destroyed completely after 60 min of sonication, so further increase in sonication time or power level cannot disintegrate the floc. There thus exist an optimum power level and sonication time for sludge disintegration [61].

Chu et al. [71] has studied the effect of sonication on the raw sludge and flocculated sludge. The size reduction in AD of the flocculated sludge after sonication was more than 50% of the sonicated raw sludge. The average surface charge of the sludge reduces due to sonication. The sonication will split the floc particles into several small particles with negatively charged surfaces. For example, due to flocculation, the ζ-potential of sludge has been increased from -14 mV to 18 mV, following sonication has reduced the ζ-potential to +4 mV, while in the original sample the ζ-potential was -14 mV [71]. Mao et al. [12] evaluated the effect of sonication on particle size reduction in primary and secondary sludge; the sludge disintegration was more in the secondary sludge compared to primary sludge. A reduction of 85% particle size has been observed in secondary and 71% in primary within 20 min of sonication. The higher reduction in the secondary sludge is expected, as it contains mostly the biomass (microbial cells), but the primary sludge consists predominantly readily settleable solids comprising fibres and less degradable cellulosic material.

The increase in sonication density has also increased the disintegration efficiency; more particles are disintegrated at higher sonication densities (73% at 4 W/mL and 60% at 2 W/mL) [12]. Bougrier et al. [47] evaluated the effect of ultrasonic treatment on particle size distribution ranging from 0.4 μm to 1000 μm of waste activated sludge (WAS) using 20 kHz frequency at different specific energy inputs. Particle sizes less than 1 μm have been observed to increase with increase in specific energy supplied. For example, E_s = 14,550 kJ/kg TS, particles of 1 μm occupied 1.5% of the whole volume, whereas they occupied 0.1% in the untreated sample. However, the volume occupied by larger particle greater than 100 μm has also been increased due to re-flocculation [69]. Akin et al. [66] studied the effect of sonication on particle size at different total solids concentration (TS) in the sludge. The size reduction was more for the lower TS sludge (2% TS content decreased by 6.5-fold at 0.67 W/mL, 240 min sonication), and a similar degree of reduction requires more ultrasonication density in the higher TS sludge (4% needs 1.03 W/mL and 6% needs 0.83 W/mL).

The effect of sonication on particle size is compared using the uniformity coefficient ($\mathrm{dp}_{60} \backslash \mathrm{dp}_{10}$), and dp_{10}. Ultrasonication has been reported to increase uniformity coefficient of sludge by 5-fold and decreases in particle size gradually with increase in specific energy [33,69]. For example, the uniformity coefficient and dp_{10} of sludge changed from 3.3 to 17 and 30.5 μm to 1.2 μm, respectively, at E_s = 7200 kJ/L [63]. The micro-flocs (<4.4 μm) shows less susceptibility to sonication than macro-flocs (>4.4 μm), as macro-flocs have the larger surface area exposed to sonication than the micro-flocs which have more binding forces, such as cells [41]. El-Hadj et al. [64] observed that with an increase in specific energy (E_s) input, the volume occupied by the smaller particles size ($\leqslant$28 μm) was more than 90%. A particle with a size larger than 4.4 μm (including, 4.4 < $d \leqslant$ 50 μm, 50 < $d \leqslant$ 125 μm and $d \geqslant$ 125 μm) exhibited more disruption than micro-flocs (<4.4 μm) [41]. The effect of particle size distribution on CST (capillary suction time) and SRF (specific resistance to filtration) was observed by Jin et al. [79] and Feng et al. [65] and they derived a strong correlation between the particle size and CST/SRF. The correlation coefficient between dp_{90} and CST was in the range of 0.8248 [79] to 0.9436 [46]. The ultrasonication breaks up small particles more effectively than larger one [46,63,64] and the particle size and energy dose were inversely related with a correlation coefficient of 0.996 at the significance level 0.01 [46].

2.4.1.2. Dewaterability of sludge. Ultrasonication has both positive and negative effects on sludge dewaterability. Lower power level

Please cite this article in press as: S. Pilli et al., Ultrasonic pretreatment of sludge: A review, Ultrason. Sonochem. (2010), doi:10.1016/j.ultsonch.2010.02.014

with less sonication time increases dewaterability, but decreases the degree of disintegration as there is no cell lysis. FitzGerald et al. [80] studied the effect of different sonication intensities on sludge dewaterability and observed a correlation between sonication and CST (suspended and total solids effects the dewaterability after sonication). Quarmby et al. [60] observed that dewaterability of the sludge decreases with an increase in ultrasonication intensity (CST increased gradually with increased in ultrasonication intensity) but anaerobic digestion of sludge has a positive effect on dewaterability, that is, dewaterability of the digested sludge increased with sonication (CST decreased). With an increase in sonication time, the dewaterability of sludge decreased gradually (CST increased from 197.4 s to 488.9 s with 60 min of sonication; the bound water content also increased by 4-folds at higher power level, 0.33 W/mL), because a greater increase in the amount of small particles formed after sonication resulted in a larger surface area for holding water [61]. Gonze et al. [69] summarized that CST of sludge decreased (dewaterability of sludge increased) with lower sonication power level and less sonication time, but with an increase in sonication time at same power level, the CST value increased. This is due to the fact that the flocs did not reduce to smaller particles at lower power levels and with less sonication time, the lower level settings favoured the sludge dewaterability.

The dewaterability of the sludge deteriorates with increase in ultrasonication intensity due to cell lysis and release of biopolymers from extracellular polymeric substances (EPS) and bacteria into aqueous phase [81]. It was stated that EPS will reduce the activated sludge dewaterability [82]. The dewaterability of sludge increases by adding flocculent to sludge before sonication [83]; by addition of flocculent will reduces the sludge water content by around 80% [84]. For example, the specific filtration resistance (SFR) of the sludge is reduced from 3.59×10^{12} m/kg to 0.43×10^{12} m/kg by addition of 100 ppm flocculent and a sonication power dose of 500 W/m^2 for 30 s. So, sonication can reduce the flocculent dose by 20–50% and increases the dewaterability of sludge [83]. In comparison to results of the above authors, Na et al. [63] observed that CST will increase initially at specific energy in the range of 0–800 kJ/L and with further increase in specific energy the CST decreases gradually. Apart from specific energy input, CST reduction is also a function of sonication time and sludge volume. As the specific energy used by previous authors were lower than the specific energy used by Na et al. [63], so dewaterability may increase with increase in specific energy at higher level.

The sludge dewaterability is favourable at CST less than 20 s [85,86]. Dewaterability of sludge can be expressed in terms of bound water, i.e., if the bound water content of the sludge increases then dewaterability decreases [87]. With increasing input power, the bound water content of the sludge increases. For example, at 0.33 W/mL, the bound water content has been reported to increase by 4-fold and thus reducing the dewaterability of the sludge [84]. Bound water content of original sludge is 3.8 kg/kg DS, and at power level of 0.11 W/mL the bound water increases to 5.9 kg/kg DS. Further at a power level of 0.33 W/mL, the bound water has increased to 11.7 kg/kg DS [84]. At higher input power, the sludge particles are disintegrated to smaller size with higher surface area causing adsorption of more water and thereby increasing the bound water [61].

The relation between the sludge dewaterability and the degree of disintegration has been evaluated by many researchers. The sludge dewaterability will increase when the degree of sludge disintegration lies between 2% and 5%. When the degree of disintegration is less than 2%, the change in sludge floc structure is very limited and when it is more than 5%, the fine particles are more which increases the bound water content [88]. Feng et al. [65] evaluated the sludge dewaterability considering the extracellular polymeric substance concentration. The optimum specific energy for the better sludge dewaterability observed is 800 kJ/kg TS, the EPS concentration is 400–500 mg/L and particle size range is 80–90 μm. When the specific energy is less than 4400 kJ/kg TS, the sludge dewaterability enhanced slightly and with dosages greater than 4400 kJ/kg TS, the dewaterability dropped significantly. At specific energy dose ranging from 0 kJ/kg to 2200 kJ/kg TS, the sludge dewaterabilty increases. Beyond the specific energy 22,000 kJ/kg the dewaterability of sludge deteriorates [65]. Further the dewaterability of sludge can be expressed in terms of CST and EPS of the sludge [62,65,89]. A correlation coefficient (R) of 0.9576 for EPS and CST, and 0.8314 for EPS and SRF has been reported [65]. Similarly, Wang et al. [62] found a good correlation coefficient of 0.9233, between EPS and CST. Houghton and Stephenson [89] have derived a quadratic relationship between EPS and CST with coefficient of determination (R^2) of 0.9687.

Feng et al. [65] evaluated the effect of specific energy on EPS release and found that with increase in specific energy, the release of EPS increases in the solution. The EPS concentration in the sludge sample increases the viscosity of the sludge [62]. Moreover, the EPS forms a thin layer on the surface of the filtering media that acts as a barrier for the water, and thus dewaterability of the sludge decreases [84] as the specific energy increases.

2.4.1.3. Settleability of sludge. The settleability of sludge changes with increase in specific energy. Feng et al. [46] have suggested a specific energy of 1000 kJ/kg TS is optimum for improving the WAS settleability. The settleability of WAS is improved, when the specific energy is less than 1000 kJ/kg (at lower specific energy the flocs are disrupted slightly, which improves the settleability), and when the specific energy is greater than 5000 kJ/kg TS, the settleability of WAS deteriorates due to complete breakdown of flocs and increase of EPS concentration in the sample [46]. The settling velocity for the activated sludge floc ranges from 5 m/h to 30 m/h, with fastest settling flocs densities ranging from 1.065 g/mL to 1.60 g/mL [90]. Chu et al. [61] concluded that ultrasonic treatment has no influence on sludge settleability, which contradicts the changes in particle size and floc structure after ultrasonication [46,63,64], and the established effects of ultrasonication on sludge [91]. The change in mass composition was evaluated by Feng et al. [46], and concluded that there is a strong correlation between specific energy input (E_s) and TDS. The optimum solids concentration range for the better sludge settleability was given by Show et al. [41], and the optimal solids content ranges from 2.3% to 3.2%. Further, Show et al. [41] observed an optimal TS range of 23,000–32,000 mg/L, and Zhang et al. [92] calculated the efficiency as "Energy efficient = (I ∗ surface area of probe ∗ sonication time)/SCOD$_{\text{increase}}$". With TS = 20,910 mg/L and 30 min sonication, Zhang et al. [92] has observed the highest energy efficiency as 166.7 kW h/kg SCOD$_{\text{increase}}$.

2.4.1.4. Microscopic examination of sludge. Ultrasonication disintegrates the sludge flocs and lysis the cell wall of the microbes. The microscopic image evaluation of microbes before and after disintegration of sludge can be used to evaluate the degree of disintegration [61]. Microscopic image evaluation provides information at cellular level of the sludge disintegrated by ultrasound [93]. The change in the structural integrity of flocs and breakdown of microbial cell walls can be observed at different sonication time [93]. With increase in sonication time complete breakdown of the flocs and cell wall will occur. For example, at 2 min of sonication the structural integrity of the flocs and filaments are significantly disrupted without appreciable destruction of bacterial cells, and at 10 min of sonication the flocs are completely disintegrated and filament-like structures with a few scattered bacterial cells, and at 30 min of sonication, more or less the complete break-up of cell walls have been observed [93]. Dewil et al. [85] have concluded

Please cite this article in press as: S. Pilli et al., Ultrasonic pretreatment of sludge: A review, Ultrason. Sonochem. (2010), doi:10.1016/j.ultsonch.2010.02.014

that ultrasound treatment reduces average size of flocs and produces abundance of separate cells and abundant short pieces of filaments (Actinomyces). In contrast to the above studies on microscopic image evaluation, Feng et al. [46] visualised that even at high energy dose 26,000 kJ/kg TS, neither the floc structure nor the microbial cells were totally disintegrated. This indicates that the ultrasonication has significant effect on microbial disruption, but the efficiency of the disruption is not clear. Still extensive future research is required is in this area to evaluate the effect of ultrasonication on microbial disruption at different specific energy input and sonication time.

2.4.1.5. Change in turbidity. The turbidity of sludge increases with increase in specific energy [33,46,63,64,75]. The lower ultrasonic frequency (20 kHz) gives high sludge disintegration efficiency, while the higher reduction in particle size during disintegration increases the turbidity of the sludge [33]. El-Hadj et al. [64] represented the effect of specific energy on aqueous phase turbidity graphically. The turbidity of the supernatant sludge decreased for specific energy less than 5000 kJ/kg TS and with specific energy greater 5000 kJ/kg TS, the turbidity increased drastically as release of micro-particles into supernatant [46]. The energy doses less than 1000 kJ/kg TS cannot disrupt large amounts of organic matter into the supernatant thus turbidity of sludge does not increased, indicating that the minimum energy required to disrupt the sludge is 1000 kJ/kg TS [46,47,69].

The evaluation of physical parameters gives a relative measurement of sludge disintegration efficiency. Still extensive research is needed between sonication parameters and their effects on physical parameters of sludge. In real field application, the changes in the physical parameters will play a major role in evaluating the sludge disintegration efficiency. The effect of ultrasonication on sludge physical parameters was evaluated by different authors as summarized in Table 3.

2.4.2. Chemical evaluation

The sludge is a complex substrate which contains various types of micro-organisms; the cell wall strength of these micro-organisms varies from each other. Chemical evaluation is 90% more quantitative and mainly focuses on sludge disintegration efficiency [9]. The “degree of disintegration (DD)” parameter was proposed by Kunz and Wagner [94] to quantify the sludge disintegration efficiency which is shown in Eq. (5) [95].

$$\mathrm{DD} = \left[\frac{\mathrm{COD_f} - \mathrm{COD_i}}{\mathrm{COD_{NaOH}} - \mathrm{COD_{NaOHo}}}\right]\left[\frac{\mathrm{COD_{NaOH^*}}}{\mathrm{COD_{homogenization}}}\right] \times 100\ (\%) \tag{5}$$

where $\mathrm{COD_f}$ is the final COD of supernatant after ultrasound treatment (mg/L), $\mathrm{COD_i}$ is the initial COD (supernatant COD, untreated) of the sample (mg/L), $\mathrm{COD_{NaOH}}$ is the COD of the supernatant at 22 h after addition of 1 M NaOH (mg/L), $\mathrm{COD_{NaOHo}}$ is the COD of supernatant just after addition of 1 M NaOH (mg/L), $\mathrm{COD_{NaOH^*}}$ is the COD of original sample right after addition of 1 M NaOH (mg/L) and $\mathrm{COD_{homogenization}}$ is the COD of original sample after homogenization.

In the above equation ($\mathrm{COD_f} - \mathrm{COD_i}$) is the soluble COD release by disintegration and ($\mathrm{COD_{NaOH}} - \mathrm{COD_{NaOHo}}$) represents the soluble COD by chemical disintegration. Chemical disintegration (NaOH) is assumed to disintegrate completely. Therefore, it is used as reference COD [9,12,33,47,48,56,61,62,69,71,94–99].

The ratio of $\mathrm{COD_{NaOH^*}}$ and $\mathrm{COD_{homogenization}}$ represents the COD of the sample before and after addition of 1 M NaOH in the ratio of 1:3.5 at 20 °C. Further Muller has modified Eq. (5) to calculate degree of disintegration ($\mathrm{DD_{COD}}$) [95] as follows:

$$\mathrm{DD_{COD}} = \left[\frac{\mathrm{COD_{ultrasound}} - \mathrm{COD_{original}}}{\mathrm{COD_{NaOH}} - \mathrm{COD_{original}}}\right] \times 100\ (\%) \tag{6}$$

The $\mathrm{COD_{ultrasound}}$ is supernatant COD of the sonicated sample (mg/L); $\mathrm{COD_{original}}$ is supernatant COD of original sample (mg/L) and $\mathrm{COD_{NaOH}}$ is the maximum COD release in the supernatant after NaOH digestion (sludge and 1 M NaOH, ratio of 1:2 for 10 min at 90 °C).

2.4.2.1. Soluble COD assessment. Ultrasonication disintegrates both cellular and extracellular matter and, organic debris and extracellular polymeric substances (EPS) of the sludge. The SCOD of sludge increases due to solubilisation of solid phase matter and increase in the concentration of organic matter and EPS in aqueous phase. Therefore, SCOD can be used as a parameter to evaluate the sludge disintegration. Apart from SCOD, ammonium nitrogen and nitrate nitrogen and EPS concentrations (polysaccharides, proteins, nucleic acids, lipids and other polymeric compounds) are also important parameters in chemical evaluation after sludge sonication. Almost all the researchers have applied SCOD as a parameter to evaluate the sludge disintegration efficiency. However, comparison of these results is very difficult, because the sludge disintegration depends on various factors like, sludge type, TS content, power supply, frequency, ultrasonic density, temperature, ultrasonication duration, sludge characteristics, etc. Ultrasonication has no effect on the total COD (TCOD) of the sludge, therefore the ratio of SCOD/TCOD represents the release of the organic matter from solid state to liquid state after ultrasonication. Tiehm et al. [33], Rai et al. [98], Bougrier et al. [47] and Nickel and Neis [100], used the degree of disintegration ($\mathrm{DD_{COD}}$) modified by Muller (modified version from Kunz and Wagner, [94]) for evaluating the disintegration efficiency as shown in Eq. (6).

Shimizu et al. [6] evaluated the solubilisation of WAS at different sonication times. To achieve a solubilisation rate of 75–80%, a minimum of 90 min ultrasonication time is required and at least 30–40 min of ultrasonication is required to get 50% solubilisation [6]. Tiehm et al. [13] reported that after 96 s of ultrasonication, the disintegration was more than 30%. With increase in sonication intensity the SCOD concentration of the WAS increases [60]. For example, at a sonication intensity of 111 W min, the increment in SCOD concentration was 9.6% and at 356 W min, the SCOD concentration increased by 15% [60]. A linear correlation was observed between degree of disintegration and applied ultrasonic intensity. The degree of disintegration was more than doubled by increasing the intensity from 6 $\mathrm{W/cm^2}$ to 18 $\mathrm{W/cm^2}$ [48]. The higher mechanical shear forces produced at higher intensities ruptures the cell walls of micro-organisms and thereby will increase the solubilisation of the COD. This in turns increases the degree of disintegration.

Further, Wang et al. [31] evaluated the effect of sonication time on sludge disintegration and found that the concentration of soluble COD, protein, and carbohydrate in the sludge or supernatant of sludge increases gradually with increase in sonication time. The ultrasonication increases the concentration of COD, protein, and carbohydrates by breaking the flocs and disrupting the cell walls of the bacteria that releases the extracellular organic compounds contained in the bacterial flocs. The dry matter of the bacteria contains about 50–60% of proteins [69]. The observed COD and carbohydrate concentration increment in the sludge with respect to increment in sonication time was relatively less when compared with protein concentration and carbohydrate concentration in the supernatant. For example, at 40 min sonication the concentration of COD, protein and carbohydrate in the supernatant was 1040 mg/L, 6000 mg/L and 1800 mg/L, respectively [31]. The ratio of sludge solubilisation is defined as the concentration of the organic substances such as protein, carbohydrates and COD in the supernatant after pretreatment to the total organic substance before pretreatment multiplied by 100 [31].

The disintegration efficiency can also be evaluated based on the release of components such as protein, polysaccharides, and DNA.

Table 3

Change in physical parameters of sludge due to ultrasonication.

Sonication condition				Physical parameters								Reference	Comments
Frequency (kHz)	Time (min)	Density (W/mL)	Power input	Particle size reduction (µm)		Settleability SVI (mL/g)		Turbidity NTU		Dewaterability (CST) (s)			
				Initial	Final	Initial	Final	Initial	Final	Initial	Final		
31	(a) 0.49 (b) 1.6	NA	3.6 kW	165	(a) 135 (b) 85	NA	NA	NA	NA	NA	NA	[13]	Particle size reduced with increase in sonication time
16–20	7.2	NA	W min (a) 111 (b) 356	NA	NA	NA	NA	NA	NA	766.1	(a) 852 (b) 904	[60]	Dewaterability deteriorated with increase in ultrasonication (US) intensity
20	120	(a) 0.11 (b) 0.33	0.110 kW	98.9	(a) 97.5 (b) 4	NA	NA	NA	NA	197.4	(a) 188 (b) 304	[51]	Particle size reduced and dewaterability deteriorated with increase in US density
20	20	0.33	NA	400	48	NA	NA	NA	NA	NA	NA	[70]	Particle size reduced with increase in US density
20	(a) 60 (b) 3 (c) 6 (d) 10	NA	0.26 kW	66.9	(a) 31.9 (b) 24.2 (c) 18.1 (d) 19.5	218	(a) 155 sonication @ 39 kJ/L (b) 125 sonication @ 156 kJ/L	NA	NA	NA	NA	[59]	The settleability of the sludge reduced with increase in specific energy input
20	20	(a) 2 (b) 3 (c) 4	0.2 kW	48 (primary sludge)	(a) 16 (b) 14 (c) 12	NA	NA	NA	NA	NA	NA	[12]	The reduction of particle size is more in secondary sludge than primary sludge at same energy input
	20	(a) 2 (b) 3 (c) 4	0.2 kW	48 (secondary sludge)	(a) 10 (b) 7 (c) 6	NA	NA	NA	NA	NA	NA		
20			0.225 kW	31.99	(a) 19.6^1 (b) 18.5^2 (c) 17.6^3 (d) 12.7^4	NA	NA	NA	NA	NA	NA	[36]	Increasing the ultrasonication resulted in smaller size particles
20	(a) 5 (b) 30	0.528	0.3–1.2 kW	NA	NA	NA	NA	NA	NA	82	(a) 344 (b) 520	[52]	The dewaterability of the is deteriorated due to sonication
NA	30	(a) 0.25 (b) 0.35 (c) 0.50	NA	NA	NA	(a) 140 (b) 130 (c) 118	(a) 65 (b) 85 (c) 70	NA	NA	NA	NA	[62]	Increasing in ultrasonication density will reduce the settleability of the sludge
20	240	(a) 0.67 (b) 1.28 (c) 2.2 (d) 3.22	2.2 kW	209 (2% TS)	(a) 32.4 (b) 28.5 (c) 22.6 (d) 18.1	NA	NA	NA	NA	NA	NA	[56]	The particle size reduction is less at higher TS content
		(a) 0.44 (b) 1.03 (c) 1.87 (d) 2.24		217 (4% TS)	(a) 185.1 (b) 36.7 (c) 33.4 (d) 38.2	NA	NA	NA	NA	NA	NA		
30	(a) 2 (b) 10	0.012	0.05–0.4 kW	65	(a) 68 (b) 41.2	NA	NA	NA	NA	NA	NA	[70]	The particle size reduction is more with increase in sonication time
20	1	(a) 0.18 (b) 0.33 (c) 0.52	1.5 kW	51	(a) 29–38 (b) 19–29 (c) 15–21	NA	NA	NA	NA	NA	NA	[63]	Increase in sonication density resulted in more particle size reduction
28		NA	kJ/L (a) 600 (b) 7200	85	4	NA	NA	35	370	50–55	(a) 68 (b) 7	[53]	The turbidity and particle size reduction increased with increase in Power input
20	NA	NA	0.07 kW	33.8	10–13.26	NA	NA	NA	With increase of E_s		Increased	[54]	Increasing the E_s input the particle size reduction
20	15	(a) 0.18 (b) 033 (c) 0.52	kW (a) 0.09	49	(a) 19 (b) 13 (c) 9	NA	NA	NA	NA	NA	NA	[41]	Increasing power input and sonication density resulted in more particle size reduction

Please cite this article in press as: S. Pilli et al., Ultrasonic pretreatment of sludge: A review, Ultrason. Sonochem. (2010), doi:10.1016/j.ultsonch.2010.02.014

			(b) 0.166 (c) 0.26										
20	1	(a) 0.18 (b) 033 (c) 0.52	kW (a) 0.09 (b) 0.166 (c) 0.26	49	(a) 32 (b) 24 (c) 13	NA	NA	NA	NA	NA	NA	[64]	Similar observations as Show et al. [41]
20	NA	E_s (a) 800 (b) 2200 (c) 4400 (d) 17,600 (e) 26,000 (f) 35,000	0.3–2 kW	94.75	(a) 88.4 (b) 61.29	NA	NA	NA	NA	94.2	(a) 83.1 (b) 89.6 (c) 112 (d) 260 (e) 339 (f) 673.4	[55]	With lower E_s input the dewaterability of the sludge increased, further increase in E_s deteriorated the sludge dewaterability
20	NA	E_s (a) 24,700 (b) 49,500 (c) 98,900 (d) 163,300	0.1 kW	NA	NA	171	(a) 118 (b) 81 (c) 47 (d) 26	171	(a) 770 (b) 1040 (c) 1520 (d) 1790	NA	NA	[66]	The turbidity and SVI increased with increase in E_s input

NA – not applicable or data not available; E_s = specific energy, kJ/kg TS.

Ultrasonication disintegrates flocs, lysis the microbial cells, which results in release of EPS and other components. The release rate of these components is not the same during the ultrasonication [81]. The protein release was higher than polysaccharides and DNA. The rate of increase was more within 20 min of sonication and beyond this the release of proteins was slower [81]. A similar trend was observed with polysaccharides and DNA. The released DNA is denatured with increase in ultrasonication density (>0.528 W/mL). In addition, increasing sonication time increases the temperature of the bulk solution and thereby denatures the released DNA [81].

Considering the performance of the disintegration and the energy input together, the optimum specific energy for sludge disintegration is 50 kJ/kg TS. A further increase in (>50 kJ/kg TS) specific energy may slow down the increment rate of protein, polysaccharides and DNA [81]. The ultrasound disintegration of sludge has been reported to increase the concentration of Ca^{+2} and Mg^{+2} into the aqueous phase. The rate of increase was higher initially and decreased with the time. The smaller particles of the sludge formed during the disintegration, absorbs the calcium and magnesium ions and decreases the concentration of Ca^{+2} and Mg^{+2} ions in the aqueous phase of the solution [81]. Ca^{+2} and Mg^{+2} released during the sonication are linked to biopolymers (protein).

The effect of total solids concentration on degree of disintegration was evaluated by Nels et al. [48]. Increase in TS, the solubilisation of SCOD increased leading to an optimum, beyond which solubilisation of SCOD decreased due to attenuation effect [12,48]. For example, the SCOD has increased from 1000 mg/L to 1800 mg/L, 4000 mg/L, 5800 mg/L and 3200 mg/L at the TS content of 0.98%, 1.7%, 2.6% and 3.6% w/v, respectively [66]. Higher solids in the liquid produces more cavitation sites and more hydro-mechanical shear forces due to implosion of more bubbles formed, and beyond the optimum concentration the homogeneous distribution of the acoustic waves are disrupted by absorption effects [48]. The degree of disintegration is lower at higher specific input, therefore the effect of ultrasonic density is more significant than specific energy input (a contradictory statement when compared with previous studies, saying that specific energy is more important than ultrasonication density for higher sludge disintegration) [62,97].

The protein concentration in the supernatant increases with increase in specific energy input at all TS content [66]. The increase of protein concentration in the supernatant was higher initially and decreased with prolong sonication [66]. At higher TS content the protein release was deteriorated due to the decreasing cavitational effect in the sludge. Show et al. [41] evaluated the effect on SCOD solubilisation with increase in ultrasonication density and TS content of the sludge. The release of SCOD in the supernatant increased with increase in ultrasonication density. At constant energy input, the optimum TS concentration range lies between 2.3% and 3.2%. To evaluate the optimal solid content range, the ultrasound disruption index D (a relationship between the disruption efficiency and the solids content) is proposed which can be calculated as:

$$D = \gamma(S/E) \tag{7}$$

where D is the ultrasound disruption index, S (mg/L) is the SCOD released in the supernatant by disruption, E (kW h/kg DS) is specific energy consumption to sonicate 1 kg dry solids of the sludge and S/E is the slope for SCOD versus specific energy input and γ is the correlation coefficient relative to sonication density, γ is regarded as 1 [41].

A new parameter, kW h/kg SCOD, has been introduced to evaluate the degree of disintegration which considers sludge characteristics and lysis efficiency [92]. Evaluating the ultrasonication of sludge with the new index parameter, the optimum operational parameters are TS in the range of 20–30 g/L, sound intensity of

158–251 W/cm^2 and sonication time of 5–15 min [92]. A positive correlation (R = 0.993, $P < 0.01$) between SCOD and the applied energy dosage was observed by Feng et al. [46]. The optimum value for complete disintegration was not found as the soluble COD went on increasing with increase in specific energy input (even at 26,000 kJ/kg TS) [46].

Tiehm et al. [33] evaluated the effect of ultrasound frequency, specific energy input and theoretical resonant cavitation bubble size on the degree of sludge disintegration. With increase in frequency the degree of disintegration decreased gradually (DD_{COD} = 80% at 41 kHz and DD_{COD} = 7% at 3217 kHz). The effect of bubble radius on degree of disintegration was calculated by considering the equation $R_r \approx 3.28 f_r^{-1}$, where R_r is the resonant bubble radius in mm and f_r is the resonance frequency in kHz [101]. The degree of the disintegration increases logarithmically with the bubble radius (>4 μm) [33].

The effect of input power and the temperature rise of the bulk solution were evaluated by Chu et al. [61] and it was observed that with an increase in power level from 0.11 W/mL to 0.33 W/mL at 120 min sonication, the total COD solubilised was 2% and 20% in the supernatant, respectively. At power level of 0.33 W/mL, the ratio of BOD/TCOD increased from 66% to 80% [61].

Considering the effect of ultrasonication on temperature, sufficient results are not available to justify an effect. The rise in temperature will leads to higher saturated vapour pressure, which will make it harder for the vapour bubble to collapse and thus decreases the intensity of cavitation [88], and hence the disintegration efficiency. Huan et al. [88] evaluated the effect of temperature on ultrasonication of sludge. The temperature of sludge increases with increase in sonication time and increases the efficiency of ultrasonication. For example, at 4 W/mL-1 min sonication with (kept at 20 °C) and without temperature control the percentage degree of disintegration was 9% for both the cases, but at 0.8 W/mL-5 min sonication, the percentage degree of disintegration was more for the uncontrolled sample (27%). For the control sample the degree of disintegration was 23%.

The effect of temperature in the bulk solution was evaluated at different power level. With increase in power level and sonication time, the temperature of the bulk solution increased gradually which resulted in an increase in the SCOD/COD ratio [61]. The effect of ultrasonic density on sludge disintegration was evaluated [62]. A low disintegration was observed due to increase in the temperature of the sludge to 65 °C in 1 h. With ultrasound density of 0.768 W/mL, the SCOD of the sludge increased from 52 mg/L to 2581 mg/L, 7509 mg/L and 8912 mg/L in 5 min, 15 min and 20 min, respectively [62]. The SCOD solubilisation rate decreased with longer sonication time.

The effect of high power and short retention time and low power and long retention time on sludge disintegration was evaluated by Gronroos et al. [97] and found no difference on SCOD released. Therefore, the optimization of the ultrasonication energy consumption is necessary for a favourable degree of disintegration. The degree of disintegration increased with an increase in dry solids (DS) content of the sludge due to the higher concentration of microbes that could be disrupted. However, the optimum DS will vary for every reactor configuration, so as to enable ultrasonic wave propagation and cell disruption. The factors limiting to achieve a maximum DS value are reactor size, transducer type, viscosity of the sludge, temperature of the sludge and polymer concentration (if polymers are added) [97]. Based on the transmutative power function model, it was concluded that power index of the sonication time was higher than that of the sonication energy density, i.e., low density and long sonication time was more efficient than the high density and less time [88]. The transmutative power function model was derived by assuming that the ultrasonic sludge disintegration as the dependent variable and its increase slows down gradually with sonication time or ultrasonication density. Generally ultrasonic sludge disintegration lies between 0 and 1. The model was expressed as:

$$DD_{COD} = 1 - (1/1 + kC^l E^m t^n) \tag{8}$$

where C is the sludge concentration (g/L); E is the ultrasonic density (W/mL); t is the sonication time (min); l, m and n are the indices of the sludge concentration, ultrasonication density and sonication time, respectively; k is the reaction rate constant depending on the ultrasonic reactor.

The solubilisation effect during in the ultrasonication in the primary and secondary sludge was evaluated [12]. The solubilisation of SCOD was higher in the secondary sludge (SCOD increased 4 times in the primary sludge and 7.7 times in the secondary sludge) and linear correlation was observed between SCOD and sonication time for primary and secondary sonicated sludge [12]. The SCOD solubilisation increases with increase in sonication densities, that is, the SCOD increased by 1.2 times, 2.3 times and 4.8 times at 2 W/mL, 3 W/mL and 4 W/mL, respectively, at the same specific energy input [12]. Bougrier et al. [47] evaluated the effect of specific energy (kJ/kg TS) on COD solubilisation. With increase in specific energy input the SCOD increased. The rate of solubilisation was higher initially between specific energy of 0–8000 kJ/kg TS, beyond which increase in specific energy reduces the rate of total solid solubilisation [47]. The soluble COD ratio (SCOD/TCOD) increased from 4% to 32% with increase in specific energy from 0 kJ/kg to 10,000 kJ/kg TS, and the optimum solubilisation occurred at a specific energy of 10,000 kJ/kg TS [47].

2.4.2.1.1. Kinetic model for ultrasonic sludge disintegration. The effect of sonication parameters and sludge characteristics on SCOD solubilisation is evaluated by developing the kinetic model. The SCOD is assumed as dependent variable and sludge concentration, pH, ultrasonic intensity, ultrasonication time and ultrasonic density as independent variables [49]. The SCOD solubilisation rate and percentage degree of disintegration can be calculated as:

$$\frac{d(SCOD+)}{dt} = k \tag{9}$$

$$\frac{d(SCOD\%)}{dt} = u \tag{10}$$

where 'k' and 'u' are given as: $k = k_0[I]^{\alpha}[pH]^{\beta}[D]^{\gamma}[C]^{\delta}$ and $u = u_0[I]^{\varphi}[pH]^{v}[D]^{\lambda}[C]^{\theta}$. '$I$' is the ultrasonication intensity, 'D' the ultrasonic density, 'C' is sludge concentration. α and φ are the influence indexes for ultrasonic density, β and v are the influence indexes of pH, λ and γ are the influence indexes for sludge concentration, δ and θ are the influence indexes for the sludge concentration, k_0 and u_0 are the intrinsic kinetics constants and are related to reaction temperature as follows:

$$\text{where } k_0 \text{ and } u_0 \text{ is defined as } k_0 = A\exp\left(\frac{-\Delta E_a}{RT}\right) \tag{11}$$

The kinetic model can be analysed using multivariable linear regression method. Wang et al. [49] showed that magnitude of each parameter on SCOD solubilisation can be arranged in the order of: sludge pH > sludge concentration > ultrasonication intensity > ultrasonic density.

The sonication time has a large effect on biomass inactivation efficiency. It is observed that the biomass inactivation occurs after 10 min of sonication [56]. Similarly, Chu et al. [61] has reported at 20 min sonication biomass inactivation using low sonication density. The above two studies reveal that inactivation of sludge is dependent on the ultrasonication density. Further, Zhang et al. [56] observed following linear relations between the degree of sludge disintegration and cell lysis with increase in ultrasonication density in the range of 0.1–1.5 W/mL.

$$\mathrm{DD_{COD}}(\%) = 38.7 \times \text{ultrasonication density (W/mL)} \quad (12)$$

$$\text{Nucleic acids (mg/L)} = 81 + 523 \times \text{power density (W/mL)} \quad (13)$$

The disintegration of sludge by ultrasonication results in increase of SCOD, supernatant protein and nucleic acids concentration [56]. The reported increase rate of supernatant protein and nucleic acids concentration is linear and can be expressed as follows:

$$\text{Nucleic acids concentration (mg/L)} = 15 + 114 \times \text{sonication time (min)} \quad (14)$$

$$\mathrm{DD_{COD}}\ (\%) = 1.2 \times \text{sonication time (min)} \quad (15)$$

$$\text{SS reduction } (\%) = 0.875 \times \text{sonication time (min)} \quad (16)$$

Evaluating the effect of disintegration and VS reduction, Zhang et al. [56] observed that VS reduction and degree of disintegration overlaps, which implies that these two parameters are interchangeable. The VS and COD represents the organic matter of the sludge, therefore the increase of the supernatant organic can be correlated with the VS reduction [56]. The degree of disintegration increases with increase in specific energy. At E_s = 10,800 kJ/L, the ratio of SCOD/TCOD has increased from 9% to 62%. Similarly, an increment in $\mathrm{DD_{COD}}$ of 80% at specific energy input of 4000 kJ/L has been reported [63]. Nels et al. [48] observed a maximum of 32% increment in $\mathrm{DD_{COD}}$ at E_s of 12,000 kJ/kg with dry solids (DS) content of 34.4 g/kg of WAS. Khanal et al. [102] have reported that optimum power input for the highest SCOD release is 35 kJ/g TS or at 3% TS content.

2.4.2.2. Protein assessment. Proteins are important building block of the bacteria that are responsible for many different functions in the living cell, for example, proteins that catalyze chemical and biochemical reactions within living cell and outside. In WAS, about 70–80% of the extracellular organic carbon is in the form of proteins and saccharides [103–107]. Wang et al. [62] have done the quantification of sludge disintegration (particularly WAS) by protein measurement. Akin et al. [66] measured the coefficient of determination (R^2) for protein increase (Δ_{protien}), $\mathrm{DD_{COD1}}$ and $\mathrm{DD_{SCOD2}}$ with respect to biogas yield (Δ_{biogas}), which are used to evaluate the sludge disintegration. For example, the combined coefficients of 0.97 for $\Delta_{\text{protien}}/\Delta_{\text{biogas}}$ were higher than that of 0.54 for $\mathrm{DD_{COD1}}/\Delta_{\text{biogas}}$, and 0.83 for $\mathrm{DD_{SCOD2}}/\Delta_{\text{biogas}}$.

Wang et al. [81] examined the release of protein in aqueous phase at different sonication times. The protein concentration has been found more predominant than DNA and polysaccharide in the aqueous phase of the sonicated sludge. The rate of release of protein was very high during the initial 20 min of sonication, while polysaccharide and DNA concentration dropped after 20 min of sonication. Feng et al. [46] have observed that the protein concentration increases with increase in energy input. An increment of 97% in the protein concentration was observed at energy input of 26,000 kJ/kg TS. Further, with high energy input the concentration of proteins is observed to increase. However, the protein measurement is not common and the calculation of ultrasound disintegration efficiency by protein measurement is not yet well accepted. Therefore, COD measurement will continue to measure the ultrasound disintegration efficiency due to its simplicity and easiness in daily operation.

2.4.2.3. NH_3 assessment. Ultrasonication increases organic nitrogen and ammonia concentration in sludge samples [9,46,47,51]. Therefore, NH_3 assessment can also be used to evaluate the degree of disintegration. Bougrier et al. [47] evaluated the effect of sonication on organic nitrogen solubilisation. At a specific energy of 15,000 kJ/kg TS, the organic nitrogen solubilisation is 40% and the maximum solubilisation occurs at a specific energy input of 10,000 kJ/kg TS [47]. With the increase of specific energy input and TS content of WAS, the release of ammonia nitrogen concentration increases [108]. The total nitrogen solubilisation increased linearly with increase in specific energy above 3600 kJ/kg TS, and a solubilisation of 19.6% was achieved at specific energy input of 108,000 kJ/kg TS [109]. The ammonia-N concentration increases due to the disintegration of bacterial cells and release of intracellular organic nitrogen into the aqueous phase, which is subsequently hydrolyzed to ammonia [51].

Feng et al. [46] have observed the changes in ammonium nitrogen and nitrogen concentration after ultrasonication at different specific energy input. The nitrate nitrogen concentration increased at ultrasonication energies higher than 5000 kJ/kg TS, while the increase of nitrate nitrogen concentration was smaller than ammonium nitrogen at similar conditions due to generation of hydroxyl radicals through acoustic cavitation [46,52]. Further the disintegration of organic nitrogen from non-biological debris is also an important contribution to ammonia nitrogen. The correlation between nitrogen data and subsequent anaerobic digestion test are required to understand the effect of ultrasonication and the release of ammonia in aqueous phase. At present, convincing conclusions are not yet reported to evaluate the disintegration efficiency of the sludge by ultrasonication [9]. The quantification of chemical parameters by various authors is tabulated in Table 4.

2.4.2.4. Biological evaluation. The ultrasonication disrupts the flocs and breaks-up the cell wall of bacteria. The breakdown of bacterial cell walls by disruption can be assessed by using biological utilization tests. Considerable amount of the WAS contains aerobic and facultative bacteria. The oxygen utilization rate (OUR) is used to characterise the microbiological activity. For example, if the OUR = 0, then all the bacterial cells are disrupted and the degree of disintegration is 100%. Thus, effectiveness of sludge disintegration by ultrasonication can be measured by OUR measurement. The term degree of inactivation ($\mathrm{DD_{OUR}}$) was introduced by Rai et al. [98], which is similar to $\mathrm{DD_{COD}}$ for evaluating the degree of disintegration. The $\mathrm{DD_{OUR}}$ is calculated using the expression of Eq. (17).

$$\mathrm{DD_{OUR}}(\%) = \left[1 - \frac{\text{OUR sonicated}}{\text{OUR original}}\right] \times 100 \quad (17)$$

where $\mathrm{OUR_{sonocated}}$ is the oxygen uptake rate of sonicated sludge, $\mathrm{OUR_{original}}$ is the oxygen uptake rate of the original sample (without sonication), and OUR can be represented by Eq. (18).

$$\mathrm{OUR} = -\frac{\mathrm{d}[\mathrm{O_2}]}{\mathrm{d}t} \quad (18)$$

The $\mathrm{DD_{OUR}}$ have been observed to increase rapidly with increase in specific energy input up to 40 kJ/g TS, beyond which it retards the $\mathrm{DD_{OUR}}$ increase rate [98]. Chu et al. [61] used the heterotrophic plate count and oxygen utilization rate (OUR) to evaluate ultrasound disintegration efficiency. With increase in sonication time, the survival ratio (ratio of viable bacteria density levels after sonication to those of original sample) of the heterotrophic bacteria decreased. For example, the survival ratio reached a value of 44% for heterotrophic bacteria and 3% for total coliform at a sonication density of 0.33 W/mL for 120 min. With increase in specific energy input, the specific oxygen uptake rate (SOUR) increases and reaches an optimum; beyond which increase in specific energy decreases the SOUR of the sludge exponentially due to inactivation of microbes [51,61,88].

At low ultrasonic densities, the floc gets disrupted, but the cell lysis does not occur, so the SOUR increases initially. A maximum of 65% degree of inactivation was observed by Akin et al. [66], and the

Please cite this article in press as: S. Pilli et al., Ultrasonic pretreatment of sludge: A review, Ultrason. Sonochem. (2010), doi:10.1016/j.ultsonch.2010.02.014

Table 4
Quantification of chemical parameters.

No.	Sludge	TS	Ultrasonic density or intensity	S_E input/frequency	Time (min)	Chemical parameters								Reference
						% COD solubilisation	DD (%)	SCOD (mg/L)		Protein (mg/L)		(%) Nitrogen solubilisation		
								Initial	Final	Initial	Final			
1	MS	NA	NA	NA	1.6	NA	30	630	2270	NA	NA	NA	NA	[13]
2	WAS	NA	NA	NA	90	75–80	NA	NA	NA	NA	NA	NA	NA	[6]
3	WAS	6–7%	111 W min	NA	NA	NA	NA	1509	1654	NA	NA	NA	NA	[60]
	WAS		356 W min	NA	NA	NA	NA	1509	1755	NA	NA	NA	NA	
4	WAS	3.3–4%	NA	NA	(a) 10 (b) 20 (c) 30 (d) 40	NA	NA	60	(a) 280 (b) 520 (c) 900 (d) 1080	110	(a) 1200 (b) 3000 (c) 5500 (d) 6000	NA	NA	[31]
5	SAS	2.3%	NA	NA	1	90	NA	100	10,000		NA	NA	NA	[110]
6	WAS	2.59%	1.8 W/cm^2	kHz (a) 41 (b) 207 (c) 360 (d) 616 (e) 1068 (f) 3217	240	NA	(a) 80 (b) 47 (c) 25 (d) 15 (e) 10 (f) 7	NA	NA	NA	NA	NA	NA	[33]
	WAS	2.59%	1.8 W/cm^2	NA	(a) 7.5 (b) 30 (c) 60 (d) 150		(a) 0.0 (b) 4.7 (c) 13.1 (d) 23.1	NA	NA	NA	NA	NA	NA	
7	WAS	38 g/L			120	18.4		2250	5000	NA	NA	NA	NA	[111]
8	AS		68.4 W/cm^2	160 kJ/L	NA	NA	9–20			NA	NA	NA	NA	[59]
9.	ES	2.45%	1.25 W/mL	kJ/kg TS (a) 3000 (b) 14,900	NA	NA		1300	(a) 2600 (b) 4050	NA	NA	NA	NA	[94]
9.	WAS	0.48%		kJ/kg TS (a) 8000 (b) 24,000 (c) 40,000 (d) 64,000	NA	NA	(a) 4.2 (b) 8 (c) 10 (d) 25	NA	NA	NA	NA	NA	NA	[91]
10	PS	% (a) 0.98 (b) 1.7 (c) 2.6 (d) 3.6	4 W/mL	NA	20	NA	NA	1020 1020	3980 (a) 2700 (b) 3000 (c) 4300 (d) 5600	NA	NA	NA	NA	[12]
	SS	% (a) 1.02 (b) 1.65 (c) 2.88 (d) 3.75	4 W/mL	NA	20	NA	NA	670 670	5260 (a) 4260 (b) 6800 (c) 9000 (d) 4400	NA	NA	NA	NA	
11	WAS	1.85%	NA	kJ/kg TS (a) 1000 (b) 15,000		(a) 8 (b) 35	(a) 14 (b) 55	NA	NA	NA	NA	NA	NA	[36]
	WAS	1.85%	NA	kJ/kg TS	NA	NA	NA	NA	NA	NA	NA	TKNs/TKN	N-NH$_4^+$/TKN	

				(a) 0 (b) 660 (c) 1355 (d) 2700 (e) 6951 (f) 14,547								(a) 3.1 (b) 8.6 (c) 11.3 (d) 15.6 (e) 29.4 (f) 44.7	(a) 2.3 (b) 4.1 (c) 4.6 (d) 5.7 (e) 11.3 (f) 18.2	
12	WAS	2.6%	5–18 W/cm^2	NA	1.06	NA	7.5–18	NA	NA	NA	NA	NA	NA	[37]
13	WAS	3%	0.768 W/mL	NA	(a) 5 (b) 15 (c) 20	NA	NA	52	(a) 2581 (b) 7509 (c) 8912	38.8	(a) 800 (b) 2300 (c) 3300	NA	NA	[93]
14	BS	NA	0.5 W/mL		(a) 10 (b) 15 (c) 20 (d) 30	NA	(a) 18 (b) 23 (c) 26 (d) 30	410	(a) 1100 (b) 1700 (c) 2700 (d) 3250	400	(a) 1800 (b) 2200 (c) 2400 (d) 2700	NA	NA	[45]
15	ES	NA	NA	1.15×10^5 kJ/kg MLVSS		44.7	NA	166	10,260	70	4701	NA	NA	[92]
16	WAS	% (a) 2 (b) 4 (c) 6	NA	5 kW s/g TS	NA	NA	NA	1.80	(a) 2.4 (b) 3.2 (c) 1.8	(a) NA (b) 0.08 (c) 0.06	(a) NA (b) 0.80 (c) 0.44	NA	NA	[56]
17	SS	2.47%	W/mL (a) 0.18 (b) 0.33 (c) 0.52	NA	1	NA	NA	523	(a) 723 (b) 890 (c) 1002	NA	NA	NA	NA	[63]
18	Sewage sludge	30.48 g/L	NA	kJ/kg TS (a) 5000 (b) 1500		(a) 6 (b) 16	(a) 12 (b) 31	NA	NA	NA	NA	NA	NA	[53]
19	WAS	2%	NA	kW s/g TS (a) 1.7 (b) 5.09 (c) 10.19 (d) 20.37 (e) 40.75	(a) 10 (b) 30 (c) 60 (d) 120 (e) 240	NA	NA	126	(a) 3800 (b) 5917 (c) 7800 (d) 8300 (e) 12,200	242	(a) 700 (b) 950 (c) 1449 (d) 1948	NA	NA	[40]
	WAS	4%	NA	kW s/g TS (a) 0.53 (b) 1.59 (c) 3.18 (d) 6.36 (e) 12.73	(a) 10 (b) 30 (c) 60 (d) 120 (e) 240	NA	NA	380	(a) 1850 (b) 3800 (c) 4400 (d) 6350 (e) 9936	138	(a) 510 (b) 740 (c) 1400 (d) 1750 (e) 1917	NA	NA	
	WAS	6%	NA	kW s/g TS (a) 0.36 (b) 1.08 (c) 2.16 (d) 4.34 (e) 8.65	(a) 10 (b) 30 (c) 60 (d) 120 (e) 240	NA	NA	420	(a) 1800 (b) 2000 (c) 3900 (d) 4500 (e) 7129	161	(a) 390 (b) 600 (c) 890 (d) 1295 (e) 1617	NA	NA	
20	WAS	14.46 g/L	NA	kJ/kg TS (a) 0 (b) 500 (c) 1000 (d) 5000 (e) 11,000 (f) 18,000 (g) 26,000	NA	NA	NA	110	(a) 110 (b) 150 (c) 180 (d) 300 (e) 790 (f) 1300 (g) 1468	538	(a) 538 (b) 600 (c) 610 (d) 820 (e) 870 (f) 900 (g) 1000	NA	$N-NH_4^+$ (a) 12 (b) 23 (c) 20 (d) 27 (e) 34 (f) 37 (g) 42	[65]

WAS – waste activated sludge; MS – mixed sludge; SAS – surplus activated sludge; AS – activated sludge; ES – excess sludge; PS – primary sludge; SS – secondary sludge; BS – biological sludge.

Please cite this article in press as: S. Pilli et al., Ultrasonic pretreatment of sludge: A review, Ultrason. Sonochem. (2010), doi:10.1016/j.ultsonch.2010.02.014

degree of inactivation based on SOUR was found to decline by 60% at a specific energy of 10 kW/g TS at 2% TS content. Similar trend was observed for higher TS concentrations (4% and 6%) in the sludge [51]. Huan et al. [88] have evaluated the relationship between the sludge microbial activity and degree of disintegration. When the sludge disintegration degree was 0–20%, microbial activity was enhanced significantly and SOUR increased by 20–40%, when the degree of disintegration was 20–40%, SOUR increased less than 20%. This indicates that some cell wall of the bacteria gets damaged prior to increase in microbial activity and when degree of disintegration reaches more than 40%, the cell lysis occurs which results in decrease of microbial activity [88]. An approximate relation between SOUR and degree of disintegration was evaluated by Huan et al. [88], which can be expressed as $DD_{SOUR} = -3.75DD_{COD}^2 + 0.75DD_{COD} + 0.21$.

3. Effects of ultrasonication on sludge degradability and methane production in anaerobic digester

The primary aim of ultrasonication is to increase the sludge biodegradability to enhance the methane production at lower HRT in anaerobic digester. Over the decades many authors have evaluated the effect of ultrasonication parameters on the sludge degradability and increased methane production. The order of pretreatment efficiency for enhancement of methane generation is: ultrasonic lysis (20 W, 9 kHz 30 min) > thermal pretreatment by autoclave (120 °C, 30 min) > thermal pretreatment with hot water (60 °C, 30 min) > freezing (−10 °C, 15 h) [15,106]. The intracellular biopolymers solubilisation and conversion to the lower molecular weight compounds of sludge through hydrolysis is a rate limiting step [4,5], and usually the hydrolysis of complex organics is catalyzed by extracellular enzymes (such as amylases, proteinases, lipases, and nucleases) [74,112,113]. Ultrasonication induces cavitation, which lysis the cell walls of microbes and releases the intracellular components into the aqueous phase. Therefore, the sonication parameters affecting cavitation will affect the sludge digestion. The increased VS reduction directly translates into increased methane generation during the anaerobic digestion and less stabilized biosolids to be disposed of.

3.1. Effects on sludge digestibility and methane

The effect of sonication density, sonication intensity and sonication time on sludge disintegration and increase in digestibility was evaluated by various authors. Shimizu et al. [6] evaluated the effect of sonication on AD of sludge in continuous digesters and found increased biogas production rate for sonicated sludge at lower HRT. For example, at 2.5 days retention time, the digestibility improved to 60% and the gas conversion efficiency improved to 40%. The gas production rate was increased till the retention time was less than 2.5 days [6]. The digestion rate of WAS and the hydrolysis rate of the biopolymers released from the WAS follows the first order kinetics, with rate constants of 0.16 day^{-1} and 1.2 day^{-1}, respectively [6]. The conversion efficiency of WAS to CH_4 is greatly improved in a two phase AD process when compared with single phase AD system [6].

Tiehm et al. [13] evaluated the effect of AD of sludge after ultrasonication by conducting batch experiments at different HRTs of 22 days, 26 days, 12 days and 8 days. The percentage VS reduction in the AD after pretreatment was on the higher side compared to the untreated sample (at 22 days). The VS reduction in untreated sample was 45.8% and in the pre-treated sample VS reduction is 50.3% at a residence time of 22 days. The VS concentration in the effluent is always 10% less than that of the conventional AD effluent [13]. Ultrasonication of sludge was observed to enhance VS reduction and biogas production in AD. For example, the biogas production for the disintegrated sludge at 22 days residence time is 36.36 L/day and at 8 days, the biogas production is 100 L/day with the volatile solids reduction of 50.3% and for the control the volatile reduction is only 45.8% at 8 days retention time. The ultrasonication of sludge increases the degradation rate of sludge, which allows for shorter retention time. For example, higher removal rate at shorter retention time, i.e., decrease of HRT from 16 days to 4 days, was observed by Nels et al. [48]. The degradation rate of VS at 4 days HRT increased by 30% as compared to the control.

The effect of sonication time and frequency of sonication was evaluated by Tiehm et al. [33] and found that with increase in pretreatment, the VS reduction in AD digester increases gradually. For example, the VS reduction in the control sample is 21.5%, and that of 30 min sonicated sample the VS reduction increased to 27.3% in the digester, an increment of 27% compared to the control. For the sample sonicated for 150 min, the VS reduction increased to 33.7%, an increment of 56.7% compared to the control [33]. The percentage biogas production of the sonicated sludge increased with increase in sonication time, and the methane percentage in the biogas increased simultaneously. Compared to the control, 8.59% increment in methane content in biogas is observed at sonication time of 150 min [33]. Evaluating the effect of frequency, with increase in ultrasound frequency the degree of disintegration decreased and the VS degradation also reduced [33].

The effect of energy input on sludge solubilisation and subsequent anaerobic digestion was evaluated by Quarmby et al. [60], and observed that the biogas production in the solubilised sludge will increase by 15% in the sonicated sludge at 356 W min compared to control. Moreover, the effect of sonication on volatile fatty acids and their effect on AD were evaluated by Quarmby et al. [60]. The volatile fatty acids (VFAs) concentration in the digester of the control sample increased from 1100 mg/L (initial VFAs in the sludge) to 1400 mg/L on the second day. After the digestion the VFA got reduced to 86 mg/L. For the sample sonicated with 356 W min, the VFAs had been increased to 1300 mg/L after sonication and in the digester, it was increased to 1800 mg/L which was 22% more compared to the control [60]. The acetic acid in the digester was analysed to evaluate the activity of the methanogenic bacteria in the digester [60]. The increment of acetic acid of the control sample was increased from 550 mg/L to 770 mg/L, only 33%, and that in the sonicated sample with 356 W min, the increment was 80%, increased from 520 mg/L to 940 mg/L [60].

The effect of sonication time on the sludge disintegration and the subsequent AD (batch test) was also evaluated by Wang et al. [31]. The percentage increment of methane increased gradually with increase in sonication time. For example, the methane amount increased by 12%, 31%, 64% and 69% on 11th day, corresponding to the sonication time of 10 min, 20 min, 30 min and 40 min, respectively [31]. Therefore, the optimum pretreatment time for enhancing the AD efficiency of WAS should be approximately 30 min [31]. The protein, carbohydrates and VFAs degradation followed a similar trend during AD. The concentration of these organic substances increased initially with 24 h (the protein concentration), 12 h (carbohydrates and VFAs digestion), thereafter, the concentration decreased gradually. The methane yield of the sonicated sludge is directly related to the increase in VFA (in particular acetate) concentration [13]. The specific methane yield decreased with increase in HRT, e.g., at 15 HRT the methane yield is 61% more compared to control whereas it was 41% higher compared to control at 25 days HRT. The increase in specific methane yield is due to increase in the net surface area of the particles and solubilisation of complex organic matter. The effect of ultrasonic density (0.2 W/mL) on methane production of the excess sludge has been evaluated [97]. The observed methane production

Please cite this article in press as: S. Pilli et al., Ultrasonic pretreatment of sludge: A review, Ultrason. Sonochem. (2010), doi:10.1016/j.ultsonch.2010.02.014

in the sonicated sludge was 8–17 times more than that of the control sample during 19 days assays. The methane production per kg of SCOD in the sample was same for 2.5 min and 10 min sonication samples, suggesting that SCOD in these samples was equally degradable despite differences in its quantities. The methane production in the anaerobic batch studies of the treated sludge (0.3 W/mL, 30 min) is 10–20% more compared to untreated sludge [97].

The effect of specific energy input on biogas production is evaluated by Bougrier et al. [47]. The biogas production increased with increase in specific energy input. For example, the biogas production for untreated sample is 20.5 mL and for the sonicated sludge, the biogas has increased to 23 mL, 25.6 mL, 25.7 mL, 31.2 mL and 32.8 mL at the specific energy of 660 kJ/kg, 1350 kJ/kg, 2700 kJ/kg, 6950 kJ/kg and 14,547 kJ/kg TS, respectively. However, at higher energies the biogas production was almost the same, and for 7000 kJ/kg and 15,000 kJ/kg TS the biogas production was same. For untreated sample about 97% of biogas is produced from the particulate matter, whereas for sonicated sludge ($E_s = 7000$ kJ/kg TS) the biogas produced from the particulate matter was 60% only [47]. The total amount of biogas increased because solids contained in the particulate part of the sludge were made soluble by ultrasonication, which are more available in the soluble fraction for the bacteria.

The effect of feed/inoculum ratio on anaerobic digestion of the sonicated sludge is evaluated by Braguglia et al. [114]. The biogas production rate increases with increasing the food/inoculum (F/I) ratio. For example, a maximum gain of 25% (biogas) was observed when the F/I ratio is 0.5 [114]. The ultrasonication pretreatment considerably enhanced the hydrolysis reaction rate constant from 0.06–0.17 day^{-1} (untreated) to 0.13–0.23 day^{-1} (sonicated) [114]. The changes in the sludge characteristics have direct relation with methane production, i.e., the rate of methane production is directly proportional to the net rate of particle size solubilisation in an anaerobic digestion [56].

The effect of sonication on hydrolysis, acidogenesis and methanogenesis and their relation was evaluated by Mao and Show [73]. The sonication appeared to be ineffective in relation to acidogenesis reaction rates (almost constant), but it provided a better buffering capacity to diminish the adverse effect of acidification [73]. The hydrolysis rate was enhanced by 19–75% for digesters fed with sonicated sludge at different sonication densities (0.18–0.52 W/mL) [73]. Therefore, the promoted biochemical reactions by sonication treatment during hydrolysis and acidogenesis was mainly attributed to the accelerated hydrolysis of complex organics, but not so much to the acidogenesis of soluble organics. The digesters fed with sonicated sludge were able to provide an amenable environment in enhancing hydrolysis of complex organics (leading to improved subsequent hydrolysis limitation) and in promoting methanogenic biomass growth, there by facilitating the initial methanogenesis limitation.

3.2. Full-scale application

Ultrasonication of sludge in enhancing COD reduction in the AD are limited to for full-scale application due to its scale-up from small scale experimental studies. Sonic Ltd., has developed a full-scale ultrasound system capable of enhancing the AD. The full-scale trials and full-scale installation around the world were done using Sonix™ (is a new technology utilizing high-powered, concentrated ultrasound for conditioning sludges). Avounmouth, Wessex water, UK, have installed an ultrasound system for treating the domestic and industrial mixed sludge (i.e., population equivalent of 1,200,000). The TS and VS reduction of the untreated sludge in the digesters was 40% and 50%, respectively, and that of sonicated sludge was 60% and 70%, respectively [25]. Similarly, Sonix™ system was installed in many plants in UK, US and Australia. Observed biogas production rate in these installations increased by 40–50% (approximately) compared with control, and the approximate VS reduction rate increased by 30–50% [25]. The effects of ultrasonication on the increment in the sludge digestibility evaluated by different authors have been summarized in Table 5.

Barber [11] has presented the details of full-scale part-stream ultrasound plants (Germany, Austria, Switzerland, Italy, and Japan), where the biogas increased by 20–50% (volume/kg fed), VS reduction improved by 20–50%, and dewatering of the sludge was improved by 3–7%. In a typical full-scale installation, the biogas production rate, VS reduction rate, and dewatering was increased by 22%, 22%, and 5–7%, respectively [11]. The energy and mass balance over a typical digester (1200 m^3, HRT – 20 days, sludge temperature 15 °C, and flow rate 200 m^3/day at 5% DS) installed with ultrasound of 2.5 W/m^2/K, operating at temperature 35 °C revealed that energy produced is more than energy consumed, i.e., 1 kW of ultrasound used will generate 7 kW of electrical energy after losses [11]. Xie et al. [115] evaluated that full-scale installation of ultrasound ('V' shape reactor volume 3.5 L, 20 kHz, 6 kW, housing five donut shaped horns stacked one over the other at a spacing of 5 cm each) for treating mixed sludge (primary and secondary sludge). In the above installation about 15–58% increase in biogas production was observed under strictly controlled conditions, with an average of 45%. Evaluating the energy balance showed that the average ratio of the net energy gain (NEG) to electric energy consumed by the ultrasound device in the operation was 2.5 (assumed every cubic meter of the methane gas generate 2.2 kW h).

4. Future perspectives

Ultrasonication of sludge has significant effects on physical, chemical and biological properties of sludge. Evaluation of physical, chemical and biological parameters will give the efficiency of ultrasonication (or disintegration of sludge by ultrasonication). The physical parameters like, particle size, turbidity, and dewaterability of sludge effected by ultrasonication and the ultrasonication parameters. The particle size of the sludge decreases with increase in ultrasonication but beyond a certain point, ultrasonication will increase the particle size due to re-flocculation. The particle size reduction varies with change in sludge characteristics and sonication parameters; therefore, the optimum ultrasonication parameters and sludge characteristics should be evaluated for each case. This can be done by mass and energy balance of the full system.

The chemical parameter evaluation is the more quantitative, and in real application, the chemical parameters (SCOD, protein concentration, ammonia concentration, VS concentration, etc.) plays a major role in evaluating the efficiency of the ultrasonication. The SCOD, protein, and ammonia concentration of the sludge increase with an increase in ultrasonication, and further increase in sonication has very little effect on these solubilisation parameters. Using high ultrasonic power with short time is as effective as low ultrasonic power with long retention time. For non-homogeneous sludge, high power and short retention and for homogenous sludge, high sonication time with low power will be more effective.

The ultrasonication of sludge is one of the emerging technologies for the sludge pretreatment to increase the biodegradability, but still extensive research on optimizing the methane yield (i.e., the net energy yield is more than energy input) is required for the full-scale application. There are more than 50 publications available, yet there is no generalised method to evaluate the efficiency of the pretreatment process. Various authors have expressed the effect of sonication parameters in different units and method of evaluation has to be standardised to compare the results of various authors. Since the cavitation is the basic phenomena of ultrasonication, the factors influencing cavitation will have a sig-

Table 5
Increment in the sludge digestibility.

No.	Type of sludge	Sonication parameters				SRT (days)	% increment with respect to control				Reference
		Power (kW)	Specific energy	Sonication time (min)	Sonication density or intensity		VS reduction	Biogas production	Methane yield	Methane content	
1	Mixed sludge	3.6	NA	1.6	NA	22	9.8	NA	NA	NA	[13]
2	WAS	NA	Batch studies		W min (a) 111 (b) 356	NA	NA	(a) 11.5 (b) 15	NA	NA	[60]
			Pilot plant studies		W min (a) 111 (b) 356	(a) 7.3 (b) 1.37	NA	NA	NA	(a) 1.01 (b) 2.60	
3	WAS	0.2	NA	(a) 10 (b) 20 (c) 30 (d) 40	NA	11	NA	NA	(a) 12 (b) 31 (c) 64 (d) 69	NA	[31]
4	SAS	9	NA	1	NA	19	NA	NA	47	5–10	[110]
	Primary sludge	NA	NA	NA	NA	(a) 15 (b) 25	NA	NA	(a) 61 (b) 41	NA	
5	WAS	NA	NA	(a) 7.5 (b) 30 (c) 60 (d) 150	NA	8	(a) 5.58 (b) 26.97 (c) 46.04 (d) 56.74	(a) −4.7 (b) 15.7 (c) 30.7 (d) 41.64	NA	(a) 1.14 (b) 4.93 (c) 7.2 (d) 8.6	[33]
6	WAS	0.047	3.4–5 kJ/g VS	1.5	NA	(a) 8 (b) 10 (c) 12	(a) −3.25 (b) −8.60 (c) 0.00	NA	NA	(a) 0.58 (b) 23 (c) −0.129	[116]
7	WAS	NA	NA	120	NA	7	89.75	20.67	19.94	NA	[111]
8	Excess sludge	Methane production assays		(a) 2.5 (b) 10	0.2 W/mL	19	NA	NA	(a) 8.5 times (b) 17 times	NA	[94]
		AD		30	0.3 W/mL	19			10–20		
9	WAS	0.025	kJ/kg TSs (a) 660 (b) 1350 (c) 2700 (d) 6950 (e) 14,547	NA	NA	NA	NA	(a) 12.9 (b) 24.87 (c) 25.36 (d) 52.19 (e) 60.00	NA	NA	[36]
10	WAS	3.6	NA	1.6	5–18 W/cm^2	(a) 18 (b) 8	(a) 31.2 (b) 41.1	(a) 13.6 (b) 22	NA	NA	[37]
11	SS	1.5	NA	1	W/mL (a) 0.18 (b) 0.33 (c) 0.52	4	(a) 14.86 (b) 22.97 (c) 25.67	(a) 148 (b) 205.7 (c) 102		(a) 3.02 (b) 10.9 (c) 12.5	[74]
12	WAS	NA	NA	1.5	10 W/cm^2	(a) 16 (b) 8	(a) 30 (b) 40.2	(a) 10.5 (b) 16.1	NA	NA	[91]

nificant effect on the ultrasonication. There is a need to evaluate the predominance of each factor (acoustic cavitation, agitation, and local heating) during the ultrasonication and their effect on degree of disintegration. The authors have evaluated the effect of ultrasonic disintegration efficiency based on the COD solubilisation and degree of disintegration, but still there is no clear picture about the effect of COD solubilisation and the specific energy input, ultrasonication density and the sonication. Also, there is a need to evaluate the correlation between these parameters. Researchers have evaluated the effect of sonication based on the VS reduction and increase in the biogas production; still uncertainties exist in judging the efficiency. Increase in viscosity of the sludge due to ultrasonication and the effect of viscosity on the performance of AD has to be evaluated.

The effect of transient cavitation (occurs when ultrasonic intensity is equal to 10 W/cm^2) and stable cavitation (non-inertial cavitation; occurs when ultrasonication intensity is between 1 W/cm^2 and 3 W/cm^2) on biodegradability of sludge and increase in methane content needs to be explored further before its successful implementation. The major disadvantage of AD is an increase in alkalinity, and therefore the release of calcium and magnesium ions during ultrasonication has a significant effect. The correlation between the amount of calcium ion and magnesium released from the sludge flocs during the sonication and their effect on AD is required for estimating the efficiency of the AD. The effects of biopolymers addition on sonication and AD has to be evaluated along with correlation with the sludge digestibility. It is also important to establish to what extent ultrasound treatment could influence each of the degradation steps for digester control and performance improvement. The sludge pH has significant effect on the disintegration efficiency; therefore, the effect of sludge pH on sludge disintegration and the AD needs to be documented. The correlation between the sludge pH and the sonication parameters and AD efficiency is necessary in full-scale application. A standard method for evaluating the efficiency of any pretreatment process is by evaluating the net energy balance and calculating the net carbon saving.

5. Conclusion

The pretreatment of the sludge by ultrasonication has a significant effect on the sludge biodegradability during the anaerobic digestion that increases biogas generation as well as percentage of methane in the biogas. Both laboratory-scale and full-scale experiments have shown a great increment in sludge reduction

Please cite this article in press as: S. Pilli et al., Ultrasonic pretreatment of sludge: A review, Ultrason. Sonochem. (2010), doi:10.1016/j.ultsonch.2010.02.014

and biogas production of the sonicated sludge in the AD. Almost 31% reduction in sludge cake can be achieved in full-scale application and also it will increase the dewaterability of sludge. The ultrasonication of sludge accelerates the conversion of complex organics into degradable substrate and it also promotes the growth of the methane-producing bacteria. The opinion of many researchers is that the effect of ultrasonic density is supposed to be more vital than the sonication time. The studies with the kinetic model have concluded that the effect of parameters is in the order of pH > sludge concentration > ultrasonication intensity > ultrasonic density. Mass and energy balance on full-scale studies showed that 1 kW of ultrasonic energy used generates about 7 kW of electrical energy after losses. Thus, higher amount of capital and operating cost can be overcome with significant reduction in the size of digesters and operating at lower HRT, which will give a significant boost to sludge management at wastewater treatment plants.

The rate of biogas production is directly proportional to the net rate of solubilisation. With increase in COD solubilisation, methane production will increase. Also, this will decrease the required HRT in the reactor, and thereby reducing the overall size of the reactor significantly. Volatile solids reduction increases with increases in ultrasonication, which will increase the degradation efficiency of the sludge in AD. Optimization of operating parameters, based upon net mass and energy balance, is of utmost important to justify the feasibility of full-scale application.

Acknowledgements

The authors are sincerely thankful to the Natural Sciences and Engineering Research Council of Canada (NSERC) (Grants A4984, Canada Research Chair) for financial support. The views or opinions expressed in this article are those of the authors and should not be construed as opinions of USEPA.

References

[1] A. Canales, A. Pareilleux, J.L. Rols, C. Goma, A. Huyard, Decreased sludge production strategy for domestic wastewater treatment, Water Sci. Technol. 30 (1994) 96–106.
[2] Canada National Inventory Report, Greenhouse Gas Sources and Sinks in Canada, 1990–2006, <www.ec.gc.ca/ghg>.
[3] S. Renzetti, Canadian Agricultural Water Use and Management. The Economics of Natural and Human Resources in Agriculture, Springer, Amsterdam, 2005.
[4] J.A. Eastman, J.F. Ferguson, Solubilization of particulate organic carbon during the acid phase of anaerobic digestion, J. Water Pollut. Control. Fed. 53 (1981) 352–366.
[5] W. Gujer, A.J.B. Zehnder, Conversion processes in anaerobic digestion, Water Sci. Technol. 12 (1983) 127–167.
[6] T. Shimizu, K. Kudo, Y. Nasu, Anaerobic waste-activated sludge digestion – a bioconversion mechanism and kinetic model, Biotechnol. Bioeng. 41 (1993) 1082–1091.
[7] S.G. Pavlostathis, J.M. Gosset, A kinetic model for anaerobic digestion of biological sludge, Biotechnol. Bioeng. 27 (1986) 1519–1530.
[8] S.G. Pavlostathis, E. Giraldo-Gomez, Kinetics of anaerobic treatment – a review, Crit. Rev. Environ. Sci. Technol. 21 (1991) 411–490.
[9] S.K. Khanal, D. Grewell, S. Sung, J. Van Leeuwen, Ultrasound applications in wastewater sludge pretreatment: a review, Crit. Rev. Environ. Sci. Technol. 37 (2007) 277–313.
[10] M.P.J. Weemaes, W.H. Verstraete, Evaluation of current wet sludge disintegration techniques, J. Chem. Technol. Biotechnol. 73 (1998) 83–92.
[11] W.P. Barber, The effects of ultrasound on sludge digestion, J. Chart. Inst. Water Environ. Manage. 19 (2005) 2–7.
[12] T. Mao, S.Y. Hong, K.Y. Show, J.H. Tay, D.J. Lee, A comparison of ultrasound treatment on primary and secondary sludges, Water Sci. Technol. 50 (2004) 91–97.
[13] A. Tiehm, K. Nickel, U. Neis, The use of ultrasound to accelerate the anaerobic digestion of sewage sludge, Water Sci. Technol. 36 (1997) 121–128.
[14] T.I. Onyeche, O. Schlafer, H. Bormann, C. Schroder, M. Sievers, Ultrasonic cell disruption of stabilised sludge with subsequent anaerobic digestion, Ultrasonics 40 (2002) 31–35.
[15] Q.H. Wang, J.C. Chen, K. Kakimoto, H.I. Ogawa, Y. Kato, Pretreatment of waste activated sludge results in enhancement of its anaerobic digestion efficiency, J. Soc. Water Environ. (Jpn.) 19 (1995) 875–882.
[16] D.E. Hughes, W.L. Nyborg, Cell disruption by ultrasound, Science 138 (1962) 108–113.
[17] H. Alliger, Ultrasonic disruption, Am. Lab. 10 (1975) 75–85.
[18] T.J. Mason, J.P. Lorimer, Applied Sonochemistry. The Uses of Power Ultrasound in Chemistry and Processing, Wiley-VCH, 2002.
[19] C.S. Hall, C.L. Dent, M.J. Scott, S.A. Wickline, High-frequency ultrasound detection of the temporal evolution of protein cross linking in myocardial tissue, IEEE Trans. Ultrason. Ferroelectr. Freq. Control. 47 (2000) 1051–1058.
[20] G. Ruecroft, D. Hipkiss, T. Ly, N. Maxted, P.W. Cains, Sonocrystallization: the use of ultrasound for improved industrial crystallization, Org. Process Res. Dev. 9 (2005) 923–932.
[21] D.R. Raichel, The Science and Applications of Acoustics, second ed., Springer, 2000. ISBN.
[22] H. Kapucu, N. Gulsoy, U. Mehmetoglu, Disruption and protein released kinetics by ultrasonication of Acetobacter peroxydan cells, Biochem. Eng. J. 5 (2000) 57–62.
[23] R.C. Sanders, B. McNeil, H.J. Finberg, S.J. Hessel, S.S. Siegelman, D.F. Adams, P.O. Alderson, H.L. Abrams, A prospective study of computed tomography and ultrasound in the detection and staging of pelvic masses, Radiology 146 (1983) 439–442.
[24] S.T.L. Harrison, Bacterial cell disruption: a key unit operation in the recovery of intracellular products, Biotechnol. Adv. 9 (1991) 217–240.
[25] F. Hogan, S. Mormede, P. Clark, M. Crane, Ultrasonic sludge treatment for enhanced anaerobic digestion, Water Sci. Technol. 50 (2004) 25–32.
[26] C.B. Scruby, L.E. Drain, Laser Ultrasonics: Techniques and Applications, Adam Hilger, 1990.
[27] D. Salerno, J. Zanetti, M. Mooney, Seismocardiography for detection of coronary artery occlusion during coronary angioplasty: a preliminary report, J. Am. Coll. Cardiol. 13 (1989) 43A.
[28] K.S. Suslick, Sonochemistry, Science 247 (1990) 1439–1445.
[29] E.B. Flint, K.S. Suslick, The temperature of cavitation, Science 253 (1991) 1397–1399.
[30] C. Petrier, Y. Jiang, M. Lamy, Ultrasound and environment: sonochemical destruction of chloromatic derivatives, Environ. Sci. Technol. 32 (1998) 1316–1318.
[31] Q. Wang, M. Kuninobu, K. Kakimoto, H.I. Ogawa, Y. Kato, Upgrading of anaerobic digestion of waste activated sludge by ultrasonic pretreatment, Bioresour. Technol. 68 (1999) 309–313.
[32] J.A. Muller, Prospects and problems of sludge pre-treatment process, Water Sci. Technol. 44 (2001) 121–128.
[33] A. Tiehm, K. Nickel, M.M. Zellhorn, U. Neis, Ultrasound waste activated sludge disintegration for improving anaerobic stabilization, Water Res. 35 (2001) 2003–2009.
[34] P.R. Gogate, A.M. Kabadi, A review of applications of cavitation in biochemical engineering/biotechnology, Biochem. Eng. 44 (2009) 60–72.
[35] L.H. Thompson, L.K. Doraiswamy, Sonochemistry: science and engineering, Ind. Eng. Chem. Res. 38 (1999) 1215–1249.
[36] Y.T. Shah, A.B. Pandit, V.S. Moholkar, Cavitation Reaction Engineering, Kluwer Academic/Plenum Publishers, New York, 1999.
[37] M.D.L.D. Castro, F.R.P. Capote, Analytical Application of Ultrasound, Elsevier, 2007. ISBN.
[38] V.S. Moholkar, I.Z. Shirgaonkar, A.B. Pandit, Cavitation and sonochemistry in the eyes of a chemical engineer, Indian Chem. Eng. 38 (1996) 81–93.
[39] J.P. Lorimer, T.J. Mason, Sonochemistry: part 1 – the physical aspects, Chem. Soc. Rev. 16 (1987) 239–274.
[40] A.D. Visscher, H.A. Langenhove, Sonochemistry of organic compounds in homogeneous aqueous oxidising systems, Ultrason. Sonochem. 5 (1998) 87–92.
[41] K.Y. Show, T. Mao, D.J. Lee, Optimization of sludge disruption by sonication, Water Res. 41 (2007) 4741–4747.
[42] T.G. Leighton, Bubble population phenomena in acoustic cavitation, Ultrason. Sonochem. 2 (1995) S123–S135.
[43] O.V. Abramov, High-Intensity Ultrasonics: Theory and Industrial Applications, Golden and Breach Science Publishers, Amesterdam, Netherlands, 1998. pp. 80–82.
[44] A. Clark, Magnetostrictive Transducer, US Patent, 1979, 4158368.
[45] Y. Chisti, Sonobioreactors: using ultrasound for enhanced microbial productivity, Trends Biotechnol. 21 (2003) 89–93.
[46] X. Feng, H.Y. Lei, J.C. Deng, Q. Yu, H.L. Li, Physical and chemical characteristics of waste activated sludge treated ultrasonically, Chem. Eng. Process. 48 (2009) 187–194 (Process Intensification).
[47] C. Bougrier, H. Carrere, J.P. Delgenes, Solubilization of waste-activated sludge by ultrasound treatment, Chem. Eng. J. 106 (2005) 163–169.
[48] U. Nels, K. Nickel, A. Tiehm, Enhancement of anaerobic sludge digestion by ultrasonic disintegration, Water Sci. Technol. 42 (2000) 73–80.
[49] F. Wang, Y. Wang, M. Ji, Mechanisms and kinetic models for ultrasonic waste activated sludge disintegration, J. Hazard. Mater. 123 (2005) 145–150.
[50] H. Kuttruff, Ultrasonic Fundamentals and Applications, Elsevier Science, Oxford, 1991.
[51] B. Akin, Waste activated sludge disintegration in an ultrasonic batch reactor, Clean – Soil, Air, Water 36 (2008) 360–365.
[52] P. Riesz, D. Berdahl, L. Christman, Free radical generation by ultrasound in aqueous and nonaqueous solutions, Environ. Health Perspect. 64 (1985) 233–252.
[53] E.J. Hart, Isotopic exchange in the sonolysis of aqueous solutions containing 14,14N2 and 15,15N2, J. Phys. Chem. 90 (1986) 5989–5991.
[54] E.J. Hart, A. Henglein, Sonolytic decomposition of nitrous oxide in aqueous solutions, J. Phys. Chem. 90 (1986) 5992–5995.

[55] K.S. Suslick, Shape selective hydrocarbon oxidation, in: C.L. Hill (Ed.), Activation and Functionalization of Alkanes, Wiley & Sons, New York, 1989.
[56] P. Zhang, G. Zhang, W. Wang, Ultrasonic treatment of biologic sludge: floc disintegration, cell lysis and inactivation, Bioresour. Technol. 98 (2007) 207–210.
[57] I.I. Hua, M.R. Hoffmann, Optimization of ultrasonic irradiation as an advanced oxidation technology, Environ. Sci. Technol. 31 (1997) 2237–2243.
[58] A. Rawle, Basic Principles of Particle Size Analysis, Technical Report, Malvern Instrument Ltd., Worcestershire, UK, 2001.
[59] A. Rawle, Basic principles of particle-size analysis, Surf. Coat. Int. 86 (2003) 58–65.
[60] J. Quarmby, J.R. Scott, A.K. Mason, G. Davies, S.A. Parsons, The application of ultrasound as a pre-treatment for anaerobic digestion, Environ. Technol. 20 (1999) 1155–1161.
[61] C.P. Chu, B.V. Chang, G.S. Liao, D.S. Jean, D.J. Lee, Observations on changes in ultrasonically treated waste-activated sludge, Water Res. 35 (2001) 1038–1046.
[62] F. Wang, M. Ji, S. Lu, Influence of ultrasonic disintegration on the dewaterability of waste activated sludge, Environ. Prog. 25 (2006) 257–260.
[63] S. Na, Y.U. Kim, J. Khim, Physiochemical properties of digested sewage sludge with ultrasonic treatment, Ultrason. Sonochem. 14 (2007) 281–285.
[64] T.B. El-Hadj, J. Dosta, R. Marquez-Serrano, J. Mata-Alvarez, Effect of ultrasound pretreatment in mesophilic and thermophilic anaerobic digestion with emphasis on naphthalene and pyrene removal, Water Res. 41 (2007) 87–94.
[65] X. Feng, J. Deng, H. Lei, T. Bai, Q. Fan, L. Zhaoxu, Dewaterability of waste activated sludge with ultrasound conditioning, Bioresour. Technol. 100 (2009) 1074–1081.
[66] B. Akin, S.K. Khanal, S. Sung, D. Grewell, J. Van-Leeuwen, Ultrasound pre-treatment of waste activated sludge, Water Sci. Technol. 6 (2006) 35–42.
[67] F. Jorand, F. Zartarian, F. Thomas, J.C. Block, J.Y. Bottero, G. Villemin, V. Urbain, J. Manem, Chemical and structural (2D) linkage between bacteria within activated sludge flocs, Water Res. 29 (1995) 1639–1647.
[68] C. Biggs, P. Lant, Identifying the Mechanisms of Activated Sludge Flocculation. Environmental Engineering Research Event, Avoca Beach, New South N, Australia, 6–9 December, 1998.
[69] E. Gonze, S. Pillot, E. Valette, Y. Gonthier, A. Bernis, Ultrasonic treatment of an aerobic activated sludge in a batch reactor, Chem. Eng. Process. 42 (2003) 965–975.
[70] Z. Guangming, Z. Panyue, C. Yanming, Ultrasonic enhancement of industrial sludge setting ability and dewatering ability, Tshinghua Sci. Technol. 11 (2006) 373–378.
[71] C.P. Chu, D.J. Lee, C.S. You, J.H. Tay, "Weak" ultrasonic pretreatment on anaerobic digestion of flocculated activated biosolids, Water Res. 36 (2002) 2681–2688.
[72] X.Q. Cao, J. Chen, Y.L. Cao, J.Y. Zhu, X.D. Hao, Experimental study on sludge reduction by ultrasound, Water Sci. Technol. 54 (2006) 87–93.
[73] T. Mao, K.Y. Show, Performance of high-rate sludge digesters fed with sonicated sludge, Water Sci. Technol. 54 (2006) 27–33.
[74] T. Mao, K.Y. Show, Influence of ultrasonication on anaerobic bioconversion of sludge, Water Environ. Res. 79 (2007) 436–441.
[75] J. Laurent, M. Casellas, M.N. Pons, C. Dagot, Flocs surface functionality assessment of sonicated activated sludge in relation with physico-chemical properties, Ultrason. Sonochem. 16 (2009) 488–494.
[76] C.F. Forster, N.J.B. Knight, D.A.J. Wase, Flocculating agents of microbial origin, Adv. Biotechnol. Prog. (1985) 211–240.
[77] J.K. Vallom, A.J. McLoughlin, Lysis as a factor in sludge flocculation, Water Res. 18 (1984) 1523–1528.
[78] L.L. Zhang, X.X. Feng, N.W. Zhu, J.M. Chen, Role of extracellular protein in the formation and stability of aerobic granules, Enzyme Microb. Technol. 41 (2007) 551–557.
[79] B. Jin, B.M. Wilen, P. Lant, Impacts of morphological, physical and chemical properties of sludge flocs on dewaterability of activated sludge, Chem. Eng. J. 98 (2004) 115–126.
[80] P.A. FitzGerald, J. Juric, C.P. Lo, G. Fleming, C.F. Forster, Sonication as a Tool in Sludge Characterisation, 15th Australian Water and Wastewater Convention, Gold Cost, Queensland, 1993.
[81] F. Wang, S. Lu, M. Ji, Components of released liquid from ultrasonic waste activated sludge disintegration, Ultrason. Sonochem. 13 (2006) 334–338.
[82] Y. Chen, H. Yang, Effect of acid and surfactant treatment on activated sludge dewatering and settling, Water Res. 35 (2001) 2615–2620.
[83] X. Yin, X.P. Lu, P.F. Han, Y.R. Wang, Ultrasonic treatment on activated sewage sludge from petro-plant for reduction, Ultrason. Sonochem. 44 (2006) 397–399.
[84] Y.G. Chen, H.Z. Yang, G.W. Gu, Effect of acid and surfactant treatment on activated sludge dewatering and settling, Water Res. 35 (2001) 2615–2620.
[85] R. Dewil, J. Baeyens, R. Goutvrind, Ultrasonic treatment of waste activated sludge, Environ. Prog. 25 (2006) 121–128.
[86] P.A. Vesilind, H.A. Davis, Using the CST device for characterising sludge dewaterability, Water Sci. Technol. 20 (1988) 203–205.
[87] X. Yin, P. Han, X. Lu, Y. Wang, A review on the dewaterability of bio-sludge and ultrasound pretreatment, Ultrason. Sonochem. 11 (2004) 337–348.
[88] L. Huan, J. Yiying, R.B. Mahar, W. Zhiyu, N. Yongfeng, Effects of ultrasonic disintegration on sludge microbial activity and dewaterability, J. Hazard. Mater. 161 (2009) 1421–1426.
[89] J.L. Houghton, T. Stephenson, Effect of influent organic content on digested sludge extracellular polymer content and dewaterability, Water Res. 36 (2002) 3620–3628.
[90] K. Sears, J.E. Alleman, J.L. Barnard, J.A. Oleszkiewicz, Density and activity characterization of activated sludge flocs, J. Environ. Eng. 132 (2006) 1235–1242.
[91] R.M. Wu, G.W. Tsou, D.J. Lee, Estimate of sludge floc permeability, Chem. Eng. J. 80 (2000) 37–42.
[92] G. Zhang, P. Zhang, J. Yang, H. Liu, Energy-efficient sludge sonication: power and sludge characteristics, Bioresour. Technol. 99 (2008) 9029–9031.
[93] S.K. Khanal, H. Isik, S. Sung, J. Avan Leeuwen, Ultrasonic conditioning of waste activated sludge for enhanced aerobic digestion, in: Proceedings of IWA Specialized Conference – Sustainable Sludge Management: State of the Art, Challenges and Perspectives, May 29–31, Moscow, Russia, 2006.
[94] P. Kunz, S. Wagner, Results and Outlooks of Investigations of Sewage Sludge Disintegration, Ergebnisse und Perspektiven aus Untersuchungen zur Klärschlammdesintegration-, awt·abwassertechnik, 1994.
[95] U. Schmitz, C.R. Berger, H. Orth, Protein analysis as a simple method for the quantitative assessment of sewage sludge disintegration, Water Res. 34 (2000) 3682–3685.
[96] J. Muller, Mechanical Disintegration of Sewage, Mechanischer Klärschlamma-ufschluß-, Schriftenreihe "Berichte aus der Verfahrenstechnik" der Fakultät für Maschinenbau und Elektrotechnik der Universität Braunschweig, Shaker Verlag, Aachen, 1996.
[97] A. Gronroos, H. Kyllonen, K. Korpijarvi, P. Pirkonen, T. Paavola, J. Jokela, J. Rintala, Ultrasound assisted method to increase soluble chemical oxygen demand (SCOD) of sewage sludge for digestion, Ultrason. Sonochem. 12 (2005) 115–120.
[98] C.L. Rai, G. Struenkmann, J. Mueller, P.G. Rao, Influence of ultrasonic disintegration on sludge growth and its estimation by respirometry, Environ. Sci. Technol. 38 (2004) 5779–5785.
[99] R. Cui, D. Jahng, Enhanced methane production from anaerobic digestion of disintegrated and deproteinized excess sludge, Biotechnol. Lett. 28 (2006) 531–538.
[100] K. Nickel, U. Neis, Ultrasonic disintegration of biosolids for improved biodegradation, Ultrason. Sonochem. 14 (2007) 450–455.
[101] F.R. Young, Cavitation, McGraw Hill, London, UK, 1989.
[102] S.K. Khanal, H. Isik, S. Sung, J. Van Leeuwen, Effects of ultrasound pretreatment on aerobic digestibility of thickened waste activated sludge, in: Proceedings of Seventh Specialized Conference on Small Water and Wastewater Systems, March 7–10, Mexico City, 2006.
[103] S. Kume, Y. Fujio, Digestion of municipal sewage sludge by a mixture of thermophilic Bacilli and their culture extract, J. Gen. Appl. Microbiol. 36 (1990) 189–194.
[104] B. Frolund, R. Palmgren, K. Keiding, P.H. Nielsen, Extraction of extracellular polymers from activated sludge using a cation exchange resin, Water Res. 30 (1996) 1749–1758.
[105] M.F. Dignac, V. Urbrain, V. Rybacki, A. Bruchet, D. Snidaro, P. Scribe, Chemical description of extracellular polymers: implication on activated sludge floc structure, Water Sci. Technol. 38 (1998) 45–53.
[106] Y.K. Kim, M.S. Kwak, W.H. Lee, J.W. Choi, Ultrasonic pretreatment for thermophilic aerobic digestion in industrial waste activated sludge treatment, Biotechnol. Bioprocess Eng. 5 (2000) 469–474.
[107] E. Neyens, J. Baeyens, R. Dewil, B.D. Heyder, Advanced sludge treatment affects extracellular polymeric substances to improve activated sludge dewatering, J. Hazard. Mater. 106 (2004) 83–92.
[108] S.K. Khanal, H. Isik, S. Sung, J. Van Leeuwen, Ultrasound pretreatment of waste activated sludge: evaluation of sludge disintegration and aerobic digestibility, in: Proceedings of IWA World Water Congress and Exhibition, September 10–14, Beijing, China, 2006.
[109] M.R. Salsabil, A. Prorot, M. Casellas, C. Dagot, Pre-treatment of activated sludge: effect of sonication on aerobic and anaerobic digestibility, Chem. Eng. J. 148 (2009) 327–335.
[110] P.B. Clark, I. Nujjoo, Ultrasonic sludge pretreatment for enhanced sludge digestion, J. CIWEM 14 (2000) 66–71.
[111] J.S. Kim, C.W. Park, T.H. Kim, Y. Lee, S. Kim, S.W. Kim, J. Lee, Effects of various pretreatment for enhanced anaerobic digestion with waste activated sludge, J. Biosci. Bioeng. 95 (2003) 271–275.
[112] A.F. Gaudy, E.T. Gaudy, Microbiology for Environmental Scientists and Engineers, McGraw-Hill, New York, 1980.
[113] H.G. Schlegel, General Microbiology, University Press, Cambridge, United Kingdom, 1993.
[114] C.M. Braguglia, G. Mininni, M.C. Tomel, E. Rolle, Effect of feed/inoculum ratio on anaerobic digestion of sonicated sludge, Water Sci. Technol. 54 (2006) 77–84.
[115] R. Xie, Y. Xing, Y.A. Ghani, K. Ooi, S. Ng, Full-scale demonstration of an ultrasonic disintegration technology in enhancing anaerobic digestion of mixed primary and thickened secondary sewage sludge, J. Environ. Eng. Sci. 6 (2007) 533–541.
[116] S. Lafitte-Trouque, C.F. Forster, The use of ultrasound and γ-irradiation as pre-treatments for the anaerobic digestion of waste activated sludge at mesophilic and thermophilic temperatures, Bioresour. Technol. 84 (2002) 113–118.

Please cite this article in press as: S. Pilli et al., Ultrasonic pretreatment of sludge: A review, Ultrason. Sonochem. (2010), doi:10.1016/j.ultsonch.2010.02.014