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# Recent Developments in the Application of Ultrasonication in Pre-Treatment of Municipal Sewage Sludge

Aleksandra Szaja<sup>1</sup>, Joanna Szulżyk-Cieplak<sup>2\*</sup>, Sylwia Łagód<sup>1</sup>, Elżbieta Kuzioła<sup>3</sup>

- <sup>1</sup> Faculty of Environmental Engineering, Lublin University of Technology, ul. Nadbystrzycka 40B, 20-618 Lublin, Poland
- <sup>2</sup> Faculty of Fundamentals of Technology , Lublin University of Technology, ul. Nadbystrzycka 38, 20-618 Lublin, Poland
- <sup>3</sup> Municipal Water Supply and Sewage Company Ltd. in Lublin, Al. Piłsudskiego 15, 20-407 Lublin, Poland
- \* Corresponding author's e-mail: j.szulzyk-cieplak@pollub.pl

# ABSTRACT

This article presents the experience in the field of using ultrasonication as a pre-treatment method of SS (sewage sludge). On the basis of a review of the literature, the effect of pretreatment of sewage sludge by US (power ultrasound) with different parameters on SS quality parameters and energy effects of the process was presented. The status of commercial applications of US as a sewage sludge pretreatment method was also presented, using the example of wastewater treatment plants in Poland. It was shown that the effective management of SS is an important technological and environmental problem in many wastewater treatment plants. Therefore, new strategies for dealing with this by-product are constantly being sought. In large wastewater treatment plants, the dominant method of stabilizing SS is AD (anaerobic digestion). However, due to the characteristics of SS, it shows low efficiency. Among the numerous strategies, US is one of the new technologies that is finding several full-scale implementations. Its application allows for solubilization of organic matter, disruption of microbial cells, as well as SS floc description and enzyme release. A number of benefits can be obtained as a result of these developments, including increased methane production, improved reaction kinetics and removal of organic matter, as well as enhanced settability and dewatering of SS, thus contributing to savings in wastewater treatment plants.

**Keywords:** sewage sludge, wastewater treatment plant, ultrasonication, sewage sludge pretreatment, sludge dewatering, sludge settability, sludge biodegradability.

## INTRODUCTION

Sewage sludge (SS) is generated as result of a number of physical, physico-chemical and biological processes taking place in a wastewater treatment plants (WWTPs). In recent years, due to expanding the sewage system and improving the wastewater treatment plant efficiency its production rate has increased gradually (Chen et al., 2022). Nevertheless, in many countries within the last two years, the systematic increase in the amount of generated SS has been stopped because of pandemic and related economic downturn. Still the global production of SS is estimated at a major value of 45 million tons of dry matter per year (Gao et al., 2020). Due to its composition e.g. presence of organic matter, heavy metals, inorganic substances, pathogens, significant humidity and its effective management, still constitute a serious concern of researchers and technologists. The main challenge is to combine several environmental and technological aspects, also taking into account the sustainable energy use (Chen et al., 2022; Ronda et al., 2023, Masłoń et al., 2020). Moreover, its proper management should be in line with the assumptions of the circular economy assume that resources are constantly used through its reuse, repair, and recycle (COM/2021/390, Rosiek, 2020). Generally, SS is considered as waste that carries a potential risk to the environment and human health (Jaromin-Gleń et al., 2017; Machnicka and Grübel, 2023). However, in

the recent years the attention of researchers has been focused on the potential of this by-product (Bagheri et al., 2023; Werle and Dudziak, 2014a). It might be a possible source of agricultural nutrients e.g. nitrogen and phosphorous a critical raw materials (Rosiek, 2020; Chojnacka et al., 2023) or valuable products e.g. biochar (Gopinath et al., 2021; Werle and Dudziak, 2014b), constructional material (Tripathi and Pal, 2023; Suchorab et al., 2016; Kosiński et el., 2023). It has been applied as animal feeds (Vethathirri et al., 2021). Another application is related with heat and energy production (Azevedo et al., 2023; Masłoń et al., 2020). Moreover, SS has been used for biofuel production, e.g. biodiesel, biohydrogen, bioethanol, and bio-oils (Roy et al., 2022; Mohamed and Li, 2023; Werle and Dudziak, 2016). All pathways are particularly important in the context of global food and energy crisis (Chojnacka et al., 2023; Liu et al., 2023).

Among various SS treatments methods anaerobic digestion (AD) has been widely applied for its stabilization. In recent years, AD has become the main unit process at large WWTPs because of its economic and environmental aspects (Lebiocka i Piotrowicz, 2012; Shin et al., 2019; Liu et al., 2021). It allows for organic matter destruction, odor reduction, pathogen inactivation while ensuring biogas and valuable digestate generation (Appels et al., 2008; Azarmanesh et al., 2023). However, despite the mentioned benefits, this method indicated particular deficiencies, mostly related with complexity and sensitivity of the AD process. It is commonly known that AD is a multistage process consisting of four steps: hydrolysis, acidogenesis, acetogenesis, and methanogenesis. Each involves various groups of microorganisms and demands particular process conditions. Only maintaining a balance between each step allows for its successful performance (Shin et al., 2019). Another problem is related with the drawbacks of SS composition; this substrate is characterised by low content of organic matter, the unfavourable carbon to nitrogen (C/N) ratio (ranging between 6-9), as well as the presence of AD inhibitors, such as heavy metals. Moreover, its low biodegradability is responsible for slow rate of hydrolysis (Grobelak et al., 2018), what is connected also with properties of significant amount of waste activated sludge (WAS) removed from main line of wastewater treatment (Szaja et al., 2015; Bieganowski et al., 2012). Each of these aspects strongly affects the efficiency of the AD, often

resulting in low methane production and process instability (Appels et al., 2008; Mata-Alvarez et al., 2014; Karki et al., 2021; Amzarmanesh et al., 2023) Therefore, various pre-treatment methods, including mechanical, chemical, biological, thermal, and their combinations have been investigated to improve SS solubilization and thus enhance the AD efficiency (Zubrowska-Sudoł, 2022; Liu et al., 2021; Grübel and Machnicka, 2020; Machnicka et al., 2019). Among these technologies, ultrasonic pre-treatment is a well-established method indicating the proven efficacy, which has been applied in many existing facilities (Zeynali et al., 2017; Zhen et al., 2017; Zieliński et al., 2018; Waclawek et al., 2019). Despite many successful applications on technical scale, there are still many technological challenges mainly relating to significant energy consumption that need to be solved (Pilli et al., 2011; Isa et al., 2020, Zhen et al., 2017). This paper presents the experience in the field of using ultrasonication as a pretreatment method of SS. The review is focused on presenting the mechanisms, recent developments, current state of commercial operations in Poland and indicating future perspective in this field.

# SEWAGE SLUDGE PRODUCTION AND ITS CHARACTERISTICS

It was evaluated that only in 2020, the total volume of treated municipal wastewater was established at level of 360-380 cubic kilometers (Giacomo and Romano, 2022; Azevedo et al., 2023). In the future perspective, a continuous increase by about 50% in 2050 in the amount of wastewater generated globally is predicted. This fact is related with progressive urbanization and developments of sewer network, mainly in populous developing countries (Giacomo and Romano, 2022). Simultaneously, with growing wastewater volume and improvements in its treatment using new technologies and sophisticated systems control, the amount of generated SS will be increased (Szelag et al., 2020; Piłat-Rożek et al., 2023; Wodecka et al., 2022). Only in China in 2019, about 39 MtDS was generated (Wei et al., 2020), while in EU it was established at a level of 13 MtDS (Bagheri et al. 2023). The increases in SS generation over the last ten years were over 2- and over 6-fold for the EU and China, respectively (Bagheri et al. 2023). However, in the few years since the pandemic and lockdown, this tendency has slowed down,

particularly in Poland. According to the Polish Central Statistical Office – GUS (2022) the total volume of wastewater as well as SS production for the last five years has remained at a relatively constant level (Fig. 1). In 2021, the total amount of generated SS at Polish municipal WWTPs was established at a level of 1025 tDS. Another visible trend concerns the downturn in investments in wastewater infrastructure. In this aspect, major growths might be observed after Poland's accession to the EU and related European funds for investments in environmental protection.

SS is recognized as main residual material produced as result of treatment process responsible for about 50% operating costs of WWTPs (Baeyens et al., 1997). Considering the application of this material in AD process, it indicates particular drawbacks related with its low bioavailability and biodegradability. The fact mostly resulted from WAS properties which consisted of microbial cells, solids, polymeric compounds as well as proteins and lipid (Bieganowski et al., 212; Drewnowski and Makinia, 2013; Drewnowski et al., 2020). All those mentioned compounds constitute a barrier and cannot be directly used by AD microbes (Abelleira-Pereira et al., 2015, Xiao, et al., 2020; Montusiewicz et al., 2010). The main limiting step in its effective application in AD is low hydrolysis rate (Xu et al., 2020; Jiang et al., 2022). Therefore, its AD is characterised by long retention times of 20–50 days and poor degradation efficacy varying between 20-50% as well as low methane production (Gil et al., 2018; Zhen et al., 2017). Hence, to improve digestion efficiency, it seems reasonable to apply a pre-treatment method to disrupt the complex structure of SS into smaller molecule compounds and release soluble organic matter that can be easily accessible for AD microbes (Zhen et al., 2017; Xiao, et al., 2020).



**Figure 1.** The situation in wastewater and sewage infrastructure in Poland in years 2000–2021: (a) municipal wastewater requiring treatment, (b) total sewage sludge generated during the year



**Figure 1. Cont.** The situation in wastewater and sewage infrastructure in Poland in years 2000–2021: (c) length of the sewage network, (d) number of municipal sewage treatment plants (GUS data, 2023)

#### PRE-TREATMENT OF SS

Generally, the pre-treatment strategies due to lower biodegradability are applied to waste activated sludge. However, in certain cases, it seems beneficial to use some techniques also to mixed or primary sludges because of enhancement pathogen deactivation and sludge quality. Thus far, thermal, ultrasonic, chemical, microwave, mechanical, biological methods and theirs combinations have been reported (Fig. 2). Though, the main goal of all techniques is boosting methane productivity and reducing solid content. For example, the results of previous studies indicate that the increase in VS (volatile solids [g/L]) reduction for selected pretreatment technologies ranged from: 6-180% for US, 23-53.1% for microwaves, 7-138% for high-pressure homogenization, 10-45% for low temperature pretreatment,

and 7-105% when using high temperature pretreatment (Neumann et al., 2016; Mitraka et al., 2022). This allows for reducing the digesters volume and hence decreasing investment and operational costs that finally might lead to improve the profitability of WWTPs (Kazimierowicz et al., 2022). However, based on the data presented in (Volschan et al. 2021), it should be noted that, in general, pretreatment methods that lead to significant improvements in biogas production also lead to high operating and maintenance costs (high-temperature thermal treatment, ultrasound, high-pressure homogenization, ozonation). Low-temperature thermal, microwave and enzyme pretreatment have lower costs and energy consumption compared to the pretreatments mentioned earlier, with significant improvements in methane production and net energy production. Enzymatic and chemical pretreatments, despite



Figure 2. The division of pre-treatment methods of sewage sludge (Nguyen et al., 2021)

lower installation costs and energy consumption, have high operating and maintenance costs. Mostly, pre-treatment strategies are employed prior the mesophilic digesters. There are several reasons for this fact. These conditions are more frequently used on a technical scale, they indicated a stable process conditions and lower energy demand (Gebreeyessus et al., 2016). Moreover, mesophilic digesters are more resistant to factors such as: temperature fluctuations, pH, and toxic substances than thermophilic ones. It should be pointed out that within pre-treatment of SS a number of changes in physicochemical properties of substrate occurred (Nguyen et al., 2021). Therefore, an implementation of pre-treatment strategy to mesophilic digesters most frequency results in a better process performance (Neumann et al., 2016; Atelge et al., 2020; Mitraka et al., 2022; Ćwiertniewicz-Wojciechowska et al., 2023).

However, each of the above-mentioned techniques presents particular limitations. Generally, physical methods are considered as demanding in terms of energy; this fact limits application on an industrial scale. Also, thermal pre-treatment might not be profitable because of relatively long time of this technology. Another significant issue is the influence of pre-treatment methods on AD stability e.g. chemical methods might disturb the pH balance in digester (Mitraka et al., 2022). It is also important to evaluate that the chosen method does not generate toxic by-products which could adversely affect the AD process (Szaja et al., 2021). The results of biological pre-treatment are difficult to predict. Additionally, the special conditions for microbial activities should be provided, that in turn can be problematic for full scale WWTPs (Zhen et al., 2017; Neumann et al., 2016). However,

several indicators should be considered before the implementation of selected method on a technical-scale (Zhen et al., 2017; Nguyen et al., 2021). Therefore, it is crucial to determine the energy consumption, estimate the costs related with e.g. dosing reagents and its influence on AD microbes. It should also apply the minimal instrumentation that can be easily operated.

### ULTRASONIC PRE-TREATMENT OF SS

Ultrasonication (US) is one of the most frequently applied methods of SS pre-treatment on a technical scale (Mitraka et al., 2022). It is recognised as most promising and highly effective method for SS disintegration (Pilli et al. 2011; Zieliński et al., 2018). This technique is included to the group of mechanical technologies that utilise a physical force to convert complex organic components into smaller particles to increase the particle surface area that can be easily accessible for AD microbes (Nguyen et al., 2021).

In this case, the cavitation phenomenon is generated through the passage of ultrasound waves with frequencies above 16 kHz in the medium. As a result of the interaction between these waves, there is an intense and sudden collapse of huge number of micro-bubbles releasing significant amounts of energy into medium, leading to extreme temperature between 4000–15 000 K and pressure conditions of up to 1.3 GPa (Gogate, 2002; Lippert et al., 2020a). These conditions promote subsequent actions e.g. impact of hydromechanical shear forces, radical formation, thermal decomposition as a result of temperature increment (Sharmila et al., 2022; Balasundaram et al., 2022; Mitraka et al., 2022). The application of US as a SS pre-treatment might lead to disruption of microorganism cells, dispergation of flocks, organic matter solubilization, enzyme release and biological activity enhancement (Neumann et al., 2016; Nguyen et al., 2021). The US might allow for a number of benefits on WWTP performance e.g. improvement settleability and dewaterability of SS, shortening of hydraulic retention time of digesters, enhancements of sludge biodegradability and methane production (Pilli et al., 2011; Ravi et al., 2023). The previous studies indicated that the best results in terms of SS might be observed at low frequencies between 20-30 kHz and high intensity field above 1.0 W cm-2 (Zielewicz, 2016). However, the cavitation effect is influenced by several operational parameters including ultrasound frequency and density, energy input, temperature as well as exposure time (Pilli et al. 2011; Delams et al., 2015). Another important factor is the type of reactor, in particular its geometry (Lippert et al., 2020). Nevertheless, the main aspect that should be considered before implementation on a technical scale is energy consumption and the related investment profitability. The influence of US pre-treatment of sewage sludge with various parameters on the sludge quality and energy effects is presented in Table 1.

US technology is recognized as highly demanding in terms of energy; therefore, the optimum US parameters and sludge characteristics should be estimated for a particular case. Only evaluation of mass and energy balances of the full system might allow for a successful implementation (Pilli et al., 2011). It should be pointed out that as a result of US application, not only increased methane production might be achieved, but also decreased pump energy demand might be obtained by lower SS viscosity (Lippert et al., 2021). Moreover, the reduced cost related with dosing of reagents might be found. No less important is the easy maintenance of US generators and their availability on the market. US also indicated a significant flexibility, it can be easily combined with other pre-treatment strategies e.g. thermal (Liu et al., 2021), hydrothermal (Zhang et al., 2023) and chemical (Wenjing et al., 2019) methods.

Currently, there are a few full-scale US installations at Polish WWTPs, e.g. in Dąbrowa Górnicza and Lublin. The first WWTPs operated 200 000 P.E. (people equivalent), therein, the US pre-treatment is applied only for WAS. The installation was provided with ULTRAWAVES

ultrasonic reactor and has been working since 2009. The recirculated digested sludge concerning 30% of the total daily TWAS flow is treated with 2 units 5kW-ULTRAWAVES US, operating 14 hours a day In this case, the implementation of this technology allowed for an improvement of the AD process resulting in boosting biogas production by 30% and enhanced degree of organics removal from 40% to 52%. Thus, the need for additional digester construction was omitted, avoiding the need for expansion and contributing to significant savings for the facility (Milanowski et al., 2013). The second WWTP serves a population of 700 000 P.E, and uses GSD (Gegen Strom Disintegration) ultrasonic disintegration technology from VTA-Technologie GmbH. Mechanical and technological start-up of the installation took place in 2015. The GSD technology of disintegration in counter-current reactors uses high-energy ultrasonic waves with a frequency of 25 kHz to disintegrate sewage sludge. Disintegration is carried out in flow-through cylindrical reactors. The system allows flexible regulation of the process parameters and optimizes the use of energy input to the sludge. Regulation of capacity and retention time allows the operator to adjust the necessary amount of energy for the proper conduct of the methane fermentation process, that is, in such a way that the energy input does not exceed the required effect. Determination of the disintegration efficiency due to the change in COD (chemical oxygen demand ) values during the tests carried out from August 2015 to March 2016 showed that it is difficult to definitively determine the efficiency of the installation. The COD increment depended on the month. This was compared with the results of dry matter content of excess sludge, and for TS = 6.1-5.7% the results were 346; 490; 538 mg/L, and for TS = 5.4-5.0% - 236; 231 mg/L (TS - total solids [g/L]). From the results, it was concluded that for the same amount of excess sludge, COD decreases along with the density of sludge. On the other hand, considering the degree of organic matter decomposition and the amount of biogas produced per unit organic matter, in August 2015, organic matter decomposition was recorded at 44.9% with a unit biogas production of 519 dm<sup>3</sup>/kgVS, and already in the following months the result of an average of 602.8  $dm^3/kgVS$  for VS = 54.3% was achieved (16%) increase). The maximum increase (21%) was 704  $dm^3/kg$  for VS = 58.8% (Trojanowska K. et al., 2017). Despite the proven effectiveness of US

	Pretreatment conditions							Effects of protrestment				
No.		Sonication parameters					Effects of pretreatment					
	Sludge parameters	P [W]	t <sub>us</sub> [s]	D <sub>us</sub> [W/mL]	F <sub>u</sub> [kHz]	ES [kWh/kg <sub>тs</sub> ]	DD <sub>cod</sub> [%]	Biogas [% incr]	Methane Yield [% incr]	TOC [% incr]	SCOD [% incr]	References.
1	Mixed sludge TS = 38.5 ± 1.2 g/L	200	0.5÷240	0.5	20	N/A	N/A	N/A	2.35÷55.79	N/A	2.9÷67.9	Şenol, 2021
2	WAS TS = 4.4 [%FM] VS = 71.7 [%FM] SCOD = 859 mgO <sub>2</sub> /L	300	N/A	N/A	25	0.1÷0.8	1.0÷22.0	N/A	1.5÷7.0	N/A	16÷890	Lippert et al., 2018
	WAS TS = 4.7 [%FM] VS = 76.6 [%FM] SCOD = 580 mgO <sub>2</sub> /L						5.0÷15.0	N/A	-2.0÷12.0	N/A	210÷520	
	WAS TS = 5.3 [%FM] VS = 73.4 [%FM] SCOD = 194 mgO <sub>2</sub> /L						1.0÷12.0	N/A	-1.0÷12.0	N/A	160÷1650	
	WAS TS = 31.9 g/L VS = 26.4 g/L SCOD =2.8 mgO <sub>2</sub> /L	100	N/A	0.1÷0.2	12		8.0÷23.0		N/A	N/A	N/A	Delmas et al., 2015
3		360		0.1÷0.72	20	1.9÷13.9	9.5-24.0	N/A				
4	Mixed sludge TS = 50.30÷50.73 mg/L VS = 36.41÷37.15mg/L COD = 602 mgO <sub>2</sub> /L	N/A	N/A	N/A	22÷27	0.2÷0.6	N/A	8.4÷11.9	N/A	N/A	N/A	Dauknys et al., 2020
5	WAS TS = 2.58 [%FM] VS = 1.77 [%FM] SCOD = 888 mgO <sub>2</sub> /L	108÷ 124	4.18÷33.54	N/A	20	1.4÷ 9.7	N/A	8.58÷31.43	N/A	N/A	28÷1361	Liazma et al., 2017
	WAS TS = 20.4+66.6 mg/L VS = 14.2+45.8 mg/L SCOD = 40+166 mgO <sub>2</sub> /L TOC = 12+192 mgO <sub>2</sub> /L	90	1800	0.136	25	• 1.5÷5.0	0.83÷18.70	- N/A	N/A	-48÷1200	73÷1298	Zielewicz, 2016
6		400	270	0.900	23		0.70÷8.20			-33÷1050	60÷1010	
		950	270	0.880	21		1.70÷5.30	10/1		-39÷5433	160÷2768	
7	WAS TS = 31.9 g/L VS = 26.4 g/L	50÷360	1440	N/A	20	1.9÷13.9	10.0÷60.0	N/A	N/A	N/A	N/A	Le et al., 2016
	WAS TS = 23 g/L VS = 13.5 g/L SCOD = 80 mgO <sub>2</sub> /L	N/A	120			0.3÷0.7	N/A	N/A	N/A	N/A	173÷491	Braguglia et. al., 2011
8			240	N/A	24	1.4		31.0÷370			173÷624	
9	Mixed sludge TS = $15 \pm 19 \text{ g/L}$ VS = $9 \pm 12 \text{ g/L}$ SCOD = $128 \text{ mgO}_2/\text{L}$	240	120÷3600	1.2	20	0.1	N/A	N/A	N/A	N/A	525÷4690	Chang et. al, 2011

Table 1. Effects of	pre-treatment of sewa	ge sludge for va	arious US p	process conditions	based on a literature review

pre-treatment achieved in both laboratory-scale and full-scale installations, there are still many areas for the future improvements. The upcoming studies should be focused on reducing the energy demand of this technology through optimization the operational parameters, such as ultrasound frequency and density as well as exposure time. Another pathway might be related with combining this method with other pre-treatment strategies or applying renewable sources of energy. Studies into new reactor configurations should also be carried out. Moreover, the influence of both physical and chemical parameters of SS on US efficiency should also be further evaluated. Thorough analyses should also concern the AD process, in particular optimization and influence of operational parameters, e.g. pH, temperature, feed/inoculum ratio, retention time and organic loading rate.

#### CONCLUSIONS

The effective management of SS is still a serious technological and environmental problem at many WWTPs. Moreover, in future perspective, incessant growth in the amount of SS generated

worldwide is projected. Therefore, new strategies for dealing with this by-product are constantly being sought. At large WWTPs, AD is the dominant method of SS stabilization. However, due to SS properties, it indicates low efficiency. The last investigations have been related to the application of various pre-treatments methods to improve SS characteristics. Among numerous strategies, US is one of the emerging technologies that finds several full-scale implementations. Its application allows for organic matter solubilisation, rupturing of microbe cells as well as description of SS flocks and releasing enzyme. As a result of these changes, a number of benefits can be obtained, e.g. increased methane production. enhanced reaction kinetics, improved organics removal, as well as upgraded settleability and dewaterability of SS, thus contributing to savings for WWTPs. However, because of high energy demand of this method, mass and energy balances of the full wastewater and sludge lines should be evaluated.

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