

Enhanced biogas production from landfill leachate by low frequency ultrasound

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Abstract

Anaerobic treatment methods have been preferred for landfill leachate which has a high organic content. However, the wastewater is also characterized as a complex content and, therefore, stability problems can occur during the operation of anaerobic reactors due to the complex structure of the wastewater. The aim of this study is to investigate the integration of an ultrasound process for leachate as a pretreatment prior to anaerobic batch reactors to increase hydrolysis rate and biogas production. After 45 min sonication, the ratio of sCOD/tCOD was increased by 32% compared to raw leachate at 400 W/l. These results showed that ultrasonic pretreatment has a significant effect on converting particulate organic matter to soluble organic matters. During the anaerobic batch tests, biogas production and methane content are increased by 33% and 15% in pretreated leachate and the raw leachate, respectively. The increases can be attributed to the improvement on biodegradability of leachate and increase in sCOD concentrations by ultrasonic pretreatment. As a conclusion, the increase in COD solubilisation by sonication enhanced biogas production and methane yield in anaerobic batch tests in leachate samples.

Keywords: landfill leachate, anaerobic treatment, ultrasound, biogas, energy.

1 Introduction

Solid waste has become one of the most important environmental problems as a result of increased population and development of industry (Bolan *et al.* [1]) and its quantity and composition changes from country to country, even city to city due to local conditions, climate and consumption patterns (Buenrostro *et al.* [2]). In Turkey, typical municipal solid waste has about 50% of organic waste, 25% of recyclable waste materials and 25% of other waste (Metin *et al.* [3]). In



developing countries, landfilling is still one of the most common ways to dispose of municipal solid waste. Although it is an economical, agreeable and commonly adopted method for solid waste management, it has nevertheless some environmental problems such as landfill leachate, methane emissions, etc. (Christian and Armour [4]). Landfill leachate which contains inorganic salts, heavy metals, high amount of biodegradable organics, and refractory compounds such as humic substances, is one of the most complex wastewaters (Li *et al.* [5]). For landfill leachate treatment, physicochemical (Kang and Hwang [6]; Ramirez and de Velasquez [7]), biological (Ağdağ and Sponza [8]) and the combinations of these methods (Wei *et al.* [9]; Aziz *et al.* [10]) have been commonly proposed in the literature. Anaerobic treatment as a biological treatment methods have been extensively used for high strength wastewaters such as landfill leachate which have significant advantages such as low sludge production, less energy requirement and biogas production (Kennedy and Lentz [11]). However, anaerobic treatment also has some disadvantages especially in reactor stabilities. Therefore, combined processes which consist of a pretreatment method prior to anaerobic treatment have been proposed for leachate, in order to enhance the degradation of complex organic matters and to obtain high biogas generation with higher methane content (He *et al.* [12]). In recent years, ultrasound treatment has been recently used for the treatment of water (Hulsman *et al.* [13]), industrial wastewater (Matouq *et al.* [14]), sludge (Tiehm *et al.* [15]) and manure (Castrillón *et al.* [16]; Elbeshbishy *et al.* [17]) for different purposes such as pre or post treatment. Especially low frequency ultrasound process has been reported to be an effective pretreatment method to enhance anaerobic treatment in terms of both organic matter solubilization and biogas production (Lehne *et al.* [18]) by enhancing the hydrolysis step. It has been reported that especially low frequency ultrasonic pretreatment improves complex organic matter degradation by reducing particle size (Nickel and Neis [19]; Bougrier *et al.* [20]). Low frequency ultrasound treatment has been used for landfill leachate in limited studies (Wnag *et al.* [21]; Neczaj *et al.* [22]) and ultrasound pretreatment prior to anaerobic treatment have been examined only in two studies for landfill leachate (Oz and Yarmitepe [23]; Bohdziewicz *et al.* [24]), so far. In these studies, it has been reported that ultrasound pretreatment combined with anaerobic processes enhance organic matter degradation and improve both biogas production rate and methane yield of biogas in landfill leachate.

With the above aforementioned, the aim of this study is to investigate the effect of ultrasound pretreatment process prior to anaerobic batch reactors for landfill leachate as a pretreatment prior to anaerobic batch reactors to increase hydrolysis rate and biodegradation so improve the anaerobic treatment efficiencies in terms of biogas production.

2 Material and method

Landfill leachate has a variable composition due to the content of the solid waste and environmental conditions. The raw leachate which is used in this study, was collected from the leachate treatment system from Istanbul Environmental



Management in Industry and Trade (ISTAC). İSTAC A.Ş stores 14,000 tons of domestic waste per day in landfill sites throughout İstanbul. In total, 3600 m³ landfill leachate is produced from this waste per day. Amount of leachate produced in Kemerburgaz/Odayeri landfill site is 2400 m³/day whereas the amount of leachate produced in Şile/Kömürcüoda landfill site is 1200 m³/day. Landfill leachate characterization is summarized in Table 1.

Table 1: Leachate characterization.

Parameter	Unit	Value
pH	-	7.38
Turbidity	NTU	1200
Total Suspended Solid	mg/l	2300
Total Chemical Oxygen Demand (tCOD)	mg/l	28500
Soluble Chemical Oxygen Demand (sCOD)	mg/l	13000
Biochemical Oxygen Demand (BOD)	mg/l	10025
Total Organic Carbon (TOC)	mg/l	7946
Total Nitrogen (TN)	mg/l	2641
Sulphate	mg/l	910
Conductivity	µS/cm	32,8
Total Phosphorus (TP)	mg/l	30.5
Nitrate	mg/l	12.5

Seed sludge samples which is used in anaerobic tests, was taken from an anaerobic expanded granular sludge bed reactor (EGSB) of a beverage factory. The concentration of the total solid, total suspended solid, total volatile solid and total volatile suspended solid of the seed sludge were monitored and is shown in Table 2.

Table 2: Seed sludge characterization.

Parameter	Unit	Value
Total Solid Matter	mg/l	69675
Total Volatile Solid	mg/l	51425
Total Suspended Solid	mg/l	35020
Total Volatile Suspended Solid	mg/l	20450

Activity of the seed sludge was determined by the Specific Methanogenic Activity (SMA) test. The SMA test unit consists of four reactors with active volume of 900 ml. A water bath was used for control the temperature ($35 \pm 2^\circ\text{C}$) and mixing (90 rpm). Biogas production during the SMA test was monitored with Milli Gas Counter (MGC) (Ritter, Germany). Biogas composition was

measured by Agilent 7820A Gas Chromatography with a thermal conductivity detector (TCD) and a molecular sieve column (HP INNOWAX 30 m x 0.25 mm and 30m x 0.50 mm). In SMA tests, four different acetate concentrations (1000-2000-3000 and 4000 mg/l) was used and the potential methane production was calculated by the formula expressed in literature (Ince *et al.* [25]). According to SMA test, maximum specific methanogenic activity of the seed sludge was determined as 398 ml CH₄/g TVS-day at optimum acetate concentration.

Complete experimental analyses were performed according to the Standard Methods (APHA [26]). Ultrasonic experimental setup was composed of a standard generator (Vibra Cell505, 500 W) equipped with a metallic probe of 1.9 cm in diameter and a supplied power of about 400 W/l at 20 kHz (Figure 1).

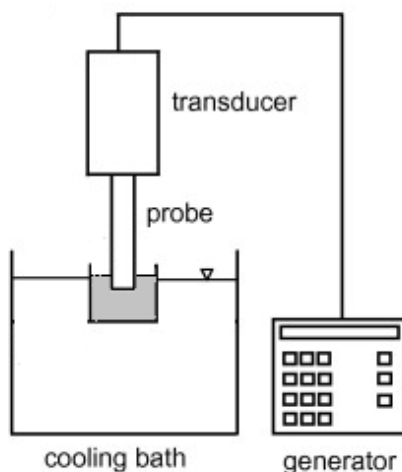


Figure 1: Ultrasonic experimental setup.

Ultrasonic treatment was applied in 250 ml of dark glass bottles which were in a water cooling bath for temperature controlling. Leachate samples were sonicated for 2, 4, 6, 8, 10, 15, 30 and 45 minutes. At the end of the ultrasonic experiments, tCOD and sCOD were analysed with Hach Lange DR 5000 Spectrophotometer according to the Standard Methods (APHA [26]). After sonification process, changes in tCOD and sCOD were monitored to investigate the effect of ultrasound process on organic matter solubilization.

The anaerobic tests of landfill leachate were performed in glass reactors with 900 ml active volume. Reactors were submerged in a water bath for temperature controlling and mixing. Raw and ultrasonically treated landfill leachate were added to reactors separately which contain seed sludge and mineral stock solution (Valcke and Verstraete [27]). In anaerobic reactors; F/M ratio were set to 0.5 and pH was set to 7. During the anaerobic tests; sCOD removal efficiencies, biogas production and methane content were monitored. sCOD removal efficiencies were determined by spectrophotometer according to standard methods. Biogas production was measured with Ritter MilliGas

Counter (Ritter, Bochum, Germany) and biogas composition especially methane content was determined with Agilent 7820A Gas Chromatography which is equipped with a thermal conductivity detector (TCD) and a molecular sieve column (HP INNOWAX 30 m x 0.25 mm and 30m x 0.50 mm).

3 Result and discussion

In this study, effects of ultrasound pretreatment on organic matter solubilization were investigated by sCOD/tCOD ratio. Raw landfill leachate samples were sonicated for up to 45 minutes (2, 4, 6, 8, 10, 15, 30 and 45 min) at 400 W/l. In low frequency sonication, while tCOD concentration was about stable (28500 ± 176 mg/l), sCOD concentration increased therefore sCOD/tCOD ratio was increased due to particulate organic matter turns to soluble forms by high cavitation energy. Table 3 shows changes in tCOD and sCOD concentration during the ultrasonic pretreatment.

Table 3: Changes in tCOD and sCOD concentration during the ultrasonic pretreatment.

Ultrasound duration, min	tCOD, mg/l	sCOD, mg/l	sCOD/tCOD
0	28500 ± 176	13000	0.46
2		13730	0.49
4		14750	0.52
6		15050	0.53
8		15750	0.55
10		16100	0.57
15		16450	0.57
30		16470	0.59
45		17400	0.61

After 45 min sonication, ratio of sCOD/tCOD increase was reached the highest value which is 32% when compare with raw leachate. Figure 2 shows the increase in ratio of sCOD/tCOD during low frequency ultrasound pretreatment.

These results showed that ultrasonic pretreatment have a significant effect on converting particulate organic matter to soluble organic matters. In literature; there are several studies which have investigated the increase of organic matter solubilisation by low frequency ultrasound for sludge and manure (Dewil *et al.* [28]; Elbeshbishy *et al.* [17]; Bougrier *et al.* [29] and Luste and Luostrarinen [30]). However, there are a few studies for wastewater (Sangave and Pandit [31], Gibson *et al.* [32] and Oz and Uzun [33]) and only two studies for landfill leachate (Oz and Yarimtepe [23]; Bohdziewicz *et al.* [24]), so far. In these studies; increase in solubilization has been reported as 35% at 600 W/l for 45 min sonication.



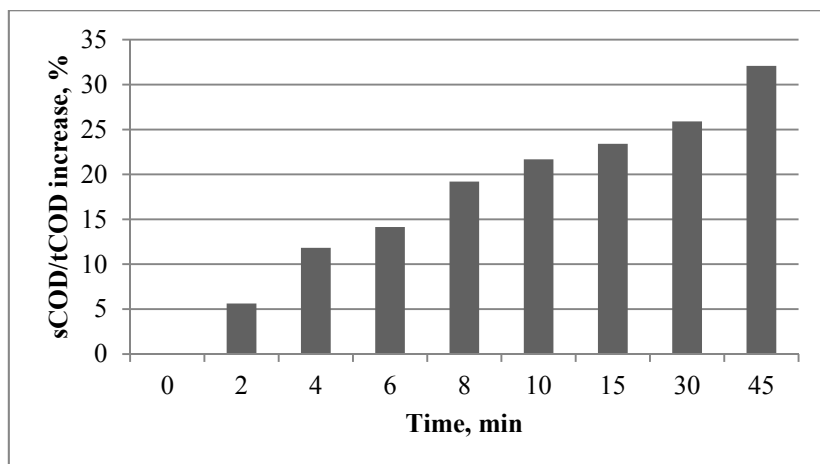


Figure 2: sCOD/tCOD increase during sonication.

After low frequency ultrasound pretreatment, anaerobic batch tests were performed with raw and pretreated landfill leachate samples for the purpose of explaining the positive effects of ultrasonic treatment prior to anaerobic treatment. After pretreatment by ultrasound, anaerobic batch tests were carried out at F/M ratio of 0.5 for both pretreated landfill leachate and raw landfill leachate as a control reactor. In anaerobic batch test; sludge samples from an anaerobic expanded granular sludge bed reactor (EGSB) of a beverage factory which have maximum specific methanogenic activity as 398 ml CH₄/g TVS-day at optimum acetate concentration, were used as seed sludge. During the anaerobic batch tests; sCOD removal efficiencies, biogas production and composition were monitored. Figure 3 shows the sCOD removal efficiencies during the anaerobic batch test. As seen from figure, ultrasonication has a favorable effect on sCOD removal efficiencies. After 45 min sonication at 400 W/l, the sCOD removal efficiency of pretreated and raw leachate was 44% and 60% in anaerobic batch tests, respectively.

Biogas production which is one of the main advantage of anaerobic treatment, was also remarkably improved by low frequency ultrasonic pretreatment during anaerobic batch tests. Total biogas production and methane yield during anaerobic batch reactors is shown in Figure 4.

In anaerobic batch tests; biogas production was determined as 341 and 453 ml while methane production of biogas were determined as 87 and 101 ml CH₄/gr VS-day for raw and pretreated landfill leachate, respectively. Therefore, in batch reactor which carried out with ultrasound pretreated leachate, 33% more biogas was obtained compared with control reactor which operated with raw leachate. During the experiments, methane content of the biogas were determined as 60% in raw landfill leachate and 69% in ultrasonically pretreated landfill leachate. Therefore, 15% increase in methane yield was obtained for ultrasonically pretreated landfill leachate. In literature, increase in methane yield has been

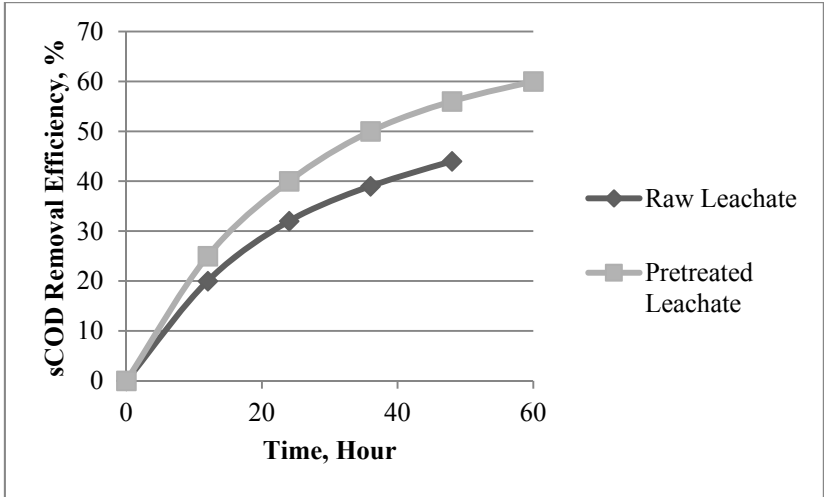


Figure 3: sCOD removal efficiencies during anaerobic batch test.

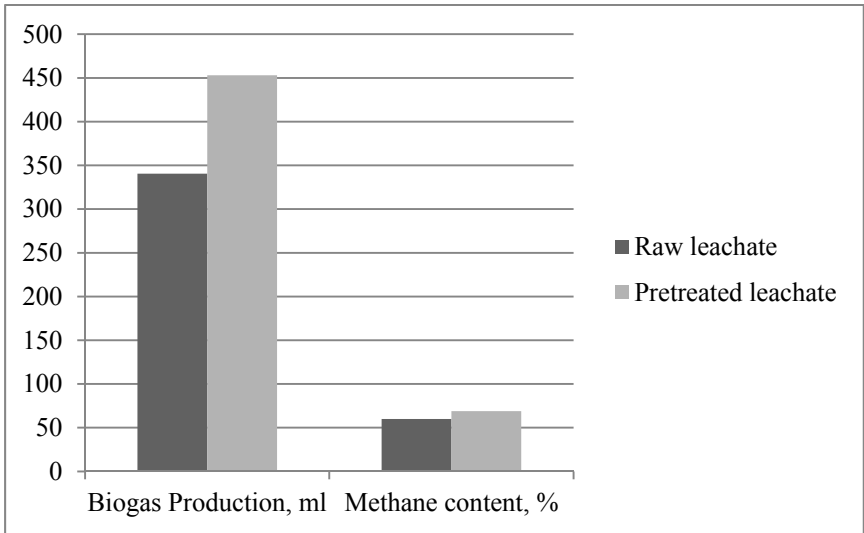


Figure 4: Total biogas production and methane content of raw and pretreated leachate.

reported between 18% and 22% for landfill leachate in various operational conditions.

Results obtained from this study showed that ultrasonic pretreatment has a positive effect on sCOD increase in pretreated landfill leachate samples and enhanced biogas production and methane yield in anaerobic batch reactors.



4 Conclusion

In this study, ultrasonication has been investigated for landfill leachate which is a strong wastewater, as a pretreatment in order to enhance anaerobic treatability for landfill leachate. Low frequency ultrasound pretreatment was performed for raw landfill leachate prior to anaerobic batch tests. After ultrasound pretreatment; it has been detected a significant increase in sCOD concentrations. Therefore, this increase in COD solubilisation by sonication enhanced biogas production and methane yield in anaerobic batch tests in landfill leachate samples when compared with the control reactor. As a conclusion, overall results showed that the ultrasound pretreatment improves anaerobic treatability of landfill leachate. There is a potential in combination of low frequency ultrasound pretreatment prior to anaerobic processes for landfill leachate treatment in terms of better reactor performance and higher biogas production with higher methane yield. This combined method can be used for other complex wastewaters to enhance anaerobic treatability. With the enhancement in anaerobic treatability, it is considered that lower reaction times and smaller reactor volumes can also be achieved.

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