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## Ultrasound assisted biogas production from landfill leachate

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#### ABSTRACT

The aim of this study is to increase biogas production and methane yield from landfill leachate in anaerobic batch reactors by using low frequency ultrasound as a pretreatment step. In the first part of the study, optimum conditions for solubilization of organic matter in leachate samples were investigated using various sonication durations at an ultrasound frequency of 20 kHz. The level of organic matter solubilization during ultrasonic pretreatment experiments was determined by calculating the ratio of soluble chemical oxygen demand (sCOD) to total chemical oxygen demand (tCOD). The sCOD/tCOD ratio was increased from 47% in raw leachate to 63% after 45 min sonication at 600 W/l. Non-parametric Friedman's test indicated that ultrasonic pretreatment has a significant effect on sCOD parameter for leachate (p < 0.05). In the second part of the study, anaerobic batch reactors were operated for both ultrasonically pretreated and untreated landfill leachate samples in order to assess the effect of sonication on biogas and methane production rate. In anaerobic batch reactor feed with ultrasonically pretreated leachate, 40% more biogas was obtained compared to the control reactor. For statistical analysis, Mann-Whitney U test was performed to compare biogas and methane production rates for raw and pretreated leachate samples and it has been found that ultrasonic pretreatment significantly enhanced biogas and methane production rates from leachate (p < 0.05) in anaerobic batch reactors. The overall results showed that low frequency ultrasound pretreatment can be potentially used for wastewater management especially with integration of anaerobic processes.

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#### 1. Introduction

Urbanization and rapid growth in population greatly accelerate consumption rates which has led to a significant increase in production rate of solid waste causing a major environmental and economical issue worldwide. Although several alternative methods including incineration, composting, recycling are proposed for controlling the solid waste problem, landfills are still one of the most predominant solid waste management methods, especially in developing countries, due to both its simplicity and economic advantages (Christian and Armour, 2000). However, the main problem with landfills is the production of leachate which is characterized with a high amount of organic matter, chlorinated organic compounds, ammonia-nitrogen, and inorganic compounds (Li et al., 2009). Due to the likely increasing restrictive environmental legislation and enforcement for protecting groundwater and surface waters, the treatment of landfill leachate is an important environmental concern. So far, different treatment options including physicochemical (Ramirez and Orta de Velasquez, 2004), biological (Ağdağ and Sponza, 2005) and the combinations of the methods (Aziz et al., 2011) have been proposed for the wastewater. However, it is a challenging issue to obtain satisfactory removal rates for complex wastewaters. This has led to an interest in testing new alternative treatment methods for landfill leachate.

Biological treatment methods especially anaerobic reactors have been extensively used for landfill leachate treatment due to significant environmental and economic benefits over aerobic treatment such as suitability to high organic loading rates, potential of biogas generation, low sludge production, less energy requirement, and reduction in  $CO_2$  emissions, (Kennedy and Lentz, 2010). However, there are still some limitations to use anaerobic processes for leachate treatment in terms of maintaining long term stability in the reactors. Therefore, combined methods comprising of pre-treatment unit and biological treatment steps have been proposed for leachate (He et al., 2007), in order to facilitate the degradation of complex organic matters and to obtain high biogas generation with higher methane content.

In recent years, ultrasound treatment has been recognized as having a potential for the treatment of water (Hulsman et al., 2010), industrial wastewater (Matouq and Al-Anber, 2007), sludge (Appels et al., 2008; Dewil et al., 2006) and manure (Elbeshbishy





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et al., 2011) for different purposes. Ultrasound is generally used to complement conventional treatment processes for pretreatment or post-treatment (Sangave and Pandit, 2004; Park et al., 2012) since required time-scale and power necessary to reach complete solubilization/mineralization of the pollutants are not feasible. Ultrasound has been extensively studied especially for sludge pretreatment before anaerobic processes to obtain better performance from anaerobic reactors by producing more bioavailable organic substrate and thereby facilitating the hydrolysis step (Muller et al., 2009). Ultrasound generates pressure waves with a high amount of energy in a medium and produces gas and vapor bubbles, growing and collapsing violently at high velocity. Bubble implosion generates regions of extreme conditions in terms of temperatures as high as 5000 K and pressures up to 100 MPa, which could lead to many physicochemical effects. The phenomenon is termed 'acoustic cavitation' which occurs especially at low frequencies between 20 and 40 kHz (Zhang et al., 2007) and produces large shear forces by jet streams during cavitation bubble implosion acting on the substances in the medium. It has been stated that the mechanism behind the solubilization process due to low frequency ultrasound relies on acoustic cavitation which causes hydro-mechanical shear forces (Zhang et al., 2007). The pretreatment process helps to disintegrate solid matter, by reducing particle size and increasing soluble organic matter fraction (Appels et al., 2008). It has been reported that anaerobic processes can be improved in terms of biogas and methane production rates as a result of ultrasonic pretreatment of waste activated sludge (Appels et al., 2008; Dewil et al., 2006). The low frequency ultrasound application due to the cavitation effects can also be an alternative for several complex industrial wastewaters such as leachate to enhance hydrolysis rate in anaerobic processes. Since pretreatment methods are used to obtain much more effective treatment, the aim of the ultrasound process as a pre-treatment option is to obtain a partial degradation of pollutants and to complement biological treatment units. Implementation of low frequency ultrasound process prior to anaerobic processes for industrial wastewaters can improve anaerobic treatability of the wastewater by enhancing solubilization of organic matter, thereby increasing biogas production and methane yield. Only few works are presently available concerning the application of low frequency ultrasound on landfill leachate (Wnag et al., 2008; Neczaj et al., 2007). To our knowledge, integration of low frequency ultrasound as a pretreatment prior to anaerobic batch tests for leachate treatment has not been reported in the literature so far.

With the above aforementioned, the aim of the proposed research is investigation of integration of ultrasound process for leachate as a pretreatment prior to anaerobic batch reactors to increase hydrolysis rate of the wastewater and to enhance the anaerobic process in terms of both biogas amount and methane yield.

#### 2. Material and methods

#### 2.1. Leachate characterization

The raw leachate was collected from the leachate treatment system from Istanbul Environmental Management in Industry and Trade (ISTAC) at the end of the February 2013. ISTAC stores 14,000 tons of domestic waste per day. The composition of leachate sample is summarized in Table 1.

#### 2.2. Source of seed sludge

Seed sludge used in anaerobic batch tests was obtained from an anaerobic expanded granular sludge bed reactor (EGSB) of a food factory in İstanbul Turkey. The concentration of the total solid

#### Table 1

Composition of landfill leachate.

Parameter	Unit	Average value <sup>*</sup> ± standard deviation
рН	-	$7.38 \pm 0.05$
Turbidity	NTU	$1200 \pm 40.82$
Total suspended solid	mg/l	2300 ± 74.54
Chemical oxygen demand (COD)	mg/l	28500 ± 324.54
Soluable chemical oxygen demand (sCOD)	mg/l	13000 ± 182.17
Biochemical oxygen demand (BOD)	mg/l	10025 ± 84.16
Conductivity	μS/cm	32.8 ± 0.35
Total organic carbon (TOC)	mg/l	7946 ± 64.62
Total nitrogen (TN)	mg/l	2641 ± 115.22
Total phosphorus (TP)	mg/l	30.5 ± 3.25
Nitrate	mg/l	$12.5 \pm 1.02$
Sulphate	mg/l	910 ± 18.39

n = 10

and total volatile solid of the sludge was about 89,000 and 70,000 mg/l respectively. Activity of the seed sludge was determined by Specific Methanogenic Activity (SMA) test as explained in Section 2.3.

#### 2.3. Specific Methanogenic Activity (SMA) Test

In this study, Specific Methanogenic Activity (SMA) test unit which is fully computerized was used to determine the activity of the seed sludge used for anaerobic batch tests. The test unit consists of four 1 L digestion reactors with active volume of 900 mL. The reactors are placed into a water bath to control the temperature within desired ranges ( $35 \pm 2 \,^{\circ}$ C). Magnetic stirrers were used for mixing at a speed of 90 rpm. Milli Gas Counter (MGC) was used for biogas measurement (Ritter, Germany). The MGC system was connected to a computer via a software (Rigamo) used to simultaneously record the gas amount. The methane content of the gas is measured by gas chromatograph. Acetate at different concentrations (1000–2000–3000 and 4000 mg/l) was used as a substrate to determine acetoclastic methanogenic activity of the sludge. The potential methane production is calculated by the formula expressed below (Ince et al., 2005):

$$SMA = (A \times B \times 24)/(C \times D)$$

SMA: Specific Methanogenic Activity (ml CH<sub>4</sub>/gTVS/d),

A: biogas production per hour (ml/h),

- B: methane content of biogas produced ( $CH_4\%$ ),
- *C*: active volume of the anaerobic test reactor (1),
- D: concentration of biomass in anaerobic test reactor (mgTVS/l).

#### 2.4. Ultrasound experiments

The experimental setup was composed of a standard generator (Vibra Cell505, 500 W) equipped with a metallic probe of 1.9 cm in diameter (Fig. 1) and a supplied power of about 200, 400 and 600 W/l at 20 kHz. The leachate sample was sonicated in a dark glass bottle which has a volume of 250 ml. During the sonication, temperature was controlled using a water-cooling bath ( $20 \,^{\circ}$ C). Samples were subjected to ultrasound for different time periods including 2, 4, 6, 8, 10, 15, 30, 45 and 60 min at different ultrasonic energy inputs (200, 400 and 600 W/l at 20 kHz). After sonication process, changes in tCOD, sCOD, total suspended solid (TSS) and turbidity parameters were monitored.

#### 2.5. Analytical methods

Complete analyses were performed according to the Standard Methods (APHA, 1998). tCOD and sCOD (after vacuum filtration through 0.45  $\mu$ m membrane) were measured with Hach Lange

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