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INFLUENCE OF ULTRASOUND FIELD ON LANDFILL LEACHATE TREATMENT BY MEANS OF ANAEROBIC PROCESS

The aim of the paper was to determine the influence of ultrasonic field on biodegradation of refractory compounds in leachate and on enhancement of treatment efficiency during anaerobic digestion process. In the first step of this investigation, the optimal ultrasound parameters, i.e. amplitude and ultrasonication time, were estimated. Thereafter the anaerobic treatment efficiency of the leachate subjected to ultrasonication and non-conditioned was assessed.

It was found that in the case of leachate ultrasonication for 300 s and at the amplitude of 14 μm, the COD removal efficiency was by 7% higher compared with that in fermentation of non-conditioned wastewater. An increase in a biogas production was also observed, while on the 8th day of the process a specific methane yield was by 22% higher compared with that of non-conditioned leachate.

Keywords: *landfill leachate, anaerobic treatment, ultrasound, anaerobic sludge granule*

1. INTRODUCTION

Storage of municipal waste in landfills is the most common method of waste disposal in Poland and in other countries. However even properly designed and exploited landfills are potentially dangerous for environment due to leachate separation. Leachate is separated due to precipitations, infiltration, surface runoff, etc.: these phenomena are responsible for infiltration of water into the landfill waste and, after saturation, generation of wastewater. Surface water and groundwater contamination is considered to be the most important effect of leachate discharge into the environment.

Some components of solid and liquid wastes deposited in landfills can be dissolved in water and then in the form of toxic and hazardous components they penetrate envi-

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ronment. So landfill leachate is comparable to complex industrial waste streams which contain both toxic organic and inorganic contaminants [1], [2].

Complete identification of all chemical compounds in leachate is difficult even if the best analytical techniqs are applied [1], [3]–[5]. Due to the presence of toxic compounds leachate was qualified as hazardous waste [6].

Since January 1998 legal obligations have been imposed on landfill exploitation. Leachate must be collected on landfill and next treated at municipal wastewater treatment plant. However, the best way is its treatment in installation localized at landfill [4].

Currently, activated sludge system is the most popular biological method used for leachate treatment. Aerobic treatment is relatively inexpensive method compared with physicochemical method and leads to the removal of biodegradable compounds, suspended solids and nitrogen compounds. Refractory compounds require some additional post-treatment processes, e.g. membrane process, adsorption on activated carbon or chemical oxidation [3].

In this paper, the influence of ultrasonic field on the biodegradation of refractory compounds in leachate and enhancement of treatment efficiency during anaerobic digestion process with anaerobic granulated sludge were investigated. Sludge granules are dense, multispecies, microbial communities and none of the individual species in the granular ecosystem is capable of degrading complex organic wastes [7], [8].

According to multilayer model the microbiological composition of granules is different in each its layer. The inner layer mainly consists of methanogens that may be the nucleation centers necessary for the initiation of granule development. H_2-producing and H_2-utilizing bacteria are dominant species in the middle layer, and mixed species predominante in the outermost layer (figure 1). To convert a target organic to methane, a spatial arrangement of methanogens and other species in anaerobic granules is essential [9]–[15].

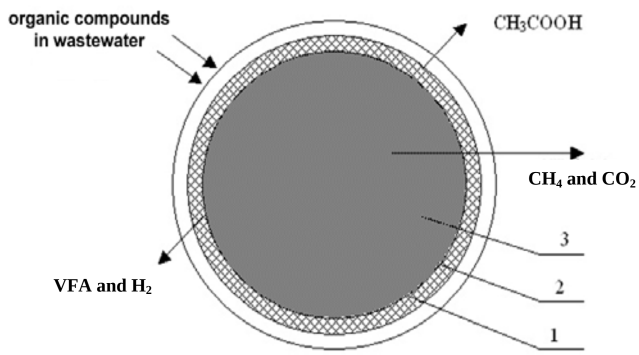

Fig. 1. Structure of anaerobic sludge granule [7]:
1 – external layer (about 14% of granule volume), 2 – middle layer (about 11% of granule volume),
3 – internal layer (about 75% of granule volume)

Anaerobic granular sludge bed technology refers to a special kind of reactor for a "high-rate" anaerobic treatment of wastewater. The concept was derived from a upward-flow anaerobic sludge blanket (UASB) reactor. Wastewater is supplied to the tank through appropriately spaced inlets. The wastewater passes upwards through an anaerobic sludge bed where the microorganisms come into contact with wastewater substrates. The resulting anaerobic degradation process typically is responsible for the production of gas (e.g. biogas containing CH_4 and CO_2). The upward motion of released gas bubbles causes hydraulic turbulence that allows reactor mixing without any mechanical equipment. At the top of the reactor, the water phase is separated from sludge solids and gas in a three-phase separator. The three-phase separator is commonly a gas cap with a settler above it [12], [13].

For several decades the ultrasonic process has been considered to be a new possibility of wastewater treating. Ultrasonic disintegration is carried out with ultrasonic field of low frequency ranging from 10 to 50 kHz and intensity over 1 W/cm^2. Sonication effect is controlled principally by the ultrasonic cavitation characteristic of an ultrasonic wave of relatively low frequency. Due to the impact of ultrasonic waves on liquid the medium is periodically compressed and rarefied. Cavitation occurs above the threshold of certain intensity, when gas bubbles are formed. They first grow in size and then violently collapse within a few microseconds. The violent collapse produces very powerful forces of hydromechanical shear in the bulk liquid surrounding the bubble. Cavitation is accomplished by high pressure gradients and extreme rise in the temperature inside the bubble. These extreme conditions can lead to the thermal destruction of compounds present in the cavitation bubbles and to the generation of very reactive hydroxyl radicals. The effects observed when cavitation occurs in aqueous solution can be summarized as: high mechanical shear stress; radical reactions, i.e. formation of OH and H radicals; chemical transformation of substances and thermal decomposition of volatile substances [16].

In the first step of this research, the optimal ultrasonic parameters, i.e. amplitude and sonication time, were estimated. In the second step, the anaerobic treatment efficiency of sonicated and non-conditioned leachate was studied.

2. MATERIALS AND METHODS

2.1. LEACHATE ANALYSIS

The municipal landfill is located closely to the urban area of the city of Częstochowa (Southern Poland) at Sobuczyna. This is an old landfill and its leachates are characterized by relatively low COD values and low ratio of BOD/COD compared with COD values characteristic of young landfills. Since 1987 municipal and non-hazardous industrial wastes have been deposited there. Currently landfill leachates are accumulated in special tanks and part of them is treated by means of reverse osmosis. Excess leachate is treated in municipal mechanical-biological wastewater treatment plant. The main characteristics of the raw leachate used in the experiments are given in table 1.

Table 1

Composition of landfill leachate from Sobuczyna

Parameter	Unit	Value
COD	Mg O_2/dm^3	3242
pH	–	8.4
Alkalinity	mg/dm^3	5800
Ammonia	mg $N–NH_4^+/dm^3$	994
Chloride	mg/dm^3	737.1

2.2. ULTRASONIC DISINTEGRATION

The disintegration of leachate was carried out in static conditions using disintegrator UD 20 with a "Sandwich" concentrator (TECHPAN Warsaw). The experimental setup was composed of an ultrasonic transducer connected to a low-frequency generator and fixed to the bottom of a 1 dm^3 reactor vessel with conditioned leachate. The ultrasonic field of 22 kHz frequency and different amplitude ranging from 8 to 16 μm was applied. During ultrasonic experiment the sonication time was changed in the range of 30–420 s.

2.3. ANAEROBIC EXPERIMENT

In order to investigate the impact of ultrasonic field on leachate degradation, an anaerobic batch test for sonicated and non-conditioned leachate was carried out. Biodegradability assays were carried out in closed Erlenmayer flasks of 300 cm^3 working capacity, which operated in batch mode.

The anaerobic leachate treatment process was conducted at the temperature of 35 °C for 4 days in the first step of this investigation and for 8 days in the second step of the experiment. The biogas produced was collected in calibrated glass cylinders filled with acidified deionized water. Seed sludge of 150 cm^3 volume was put into flask and diluted with waste of 150 cm^3 volume, consisting of synthetic waste and leachate (20 % v/v). Then the content of flask was sonicated at different parameters, i.e. amplitude and sonication time.

In the flask, the sludge concentration of 45.2 g/dm^3 (34.9 g of organic matter per

1 dm^3 and 10.3 g of mineral matter per 1 dm^3) was maintained.

Anaerobic reactor operated at organic loading rate of 1.26 kg COD/m^3·d and sludge loading rate of 0.036 kg COD/kg VSS·d. The value of COD in synthetic waste changed in the range of 2400–2550 mg O_2/dm^3.

Chemical composition of synthetic waste being prepared according to PN-72/C-04550 was as follows:

- mineral component I, K_2HPO_4 (6 g/dm^3),
- mineral component II, $CaCl_2 \cdot 7H_2O$ (0.6 g/dm^3), KH_2PO_4 (6 g/dm^3), $MgSO_4 \cdot 7H_2O$ (2.6 g/dm^3), NaCl (12 g/dm^3), NH_4Cl (6 g/dm^3),
- glucose (2.343 g/dm^3),
- sodium hydrogen carbonate (2.5 g/dm^3).

COD removal efficiency was calculated as follows:

$$X = \frac{a-b}{a} \times 100\%, \tag{1}$$

where:

a – COD in raw wastewater before digestion process [mg O_2/dm^3],

b – COD in wastewater after four days of digestion [mg O_2/dm^3].

Chemical oxygen demand was analyzed colorimetrically using tests and photometer of the HACH firm (DR 4000).

Synthetic wastewater containing 20% (v/v) of leachate that was not conditioned by ultrasounds was digested in four flask. In another four flasks, leachate was subjected to anaerobic digestion under the same conditions, but before biochemical stabilization leachate was ultrasonicated for 300 s at 14 μm amplitude. The efficiency of both processes was evaluated based on the changes in such parameters as: COD, pH, alkalinity, volatile fatty acids (VFA), chloride, ammonia and biogas production. Methane specific yield was calculated according to the following formula:

$$Y = \frac{V_b}{a} \ [\mathrm{dm^3\,g^{-1}COD_{removed}}], \tag{2}$$

where:

V_b – biogas production per day [dm^3/d],

a – COD removal per day [g/d].

3. RESULTS

In the first step of this investigation, the dependence of ultrasonic parameters, i.e. the amplitude and time of ultrasonication, on COD of ultrasonicated leachate was estimated. The results obtained during the first part of this research are shown in figure 2.

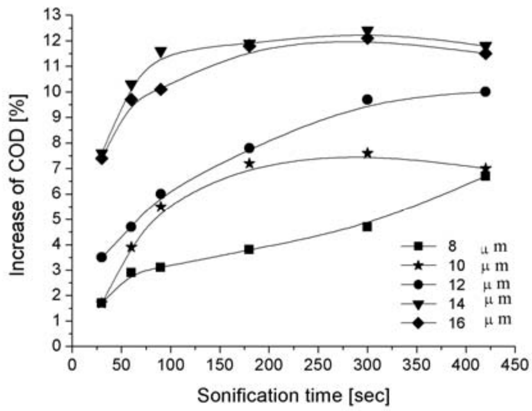

Fig. 2. An increase in COD in landfill leachate versus amplitude of ultrasonic field and the time of ultrasonication

Chemical oxygen demand in raw leachate was 3429 mg O_2/dm^3. The highest increase in the value of COD at all times of ultrasonication was measured for the amplitudes of 14 μm and 16 μm. Changing the time of ultrasonication in the range of 30–420 s we obtained a maximum increase in COD value in conditioned leachate; 12.4% (14 μm, 300 s) and 12.1% (16 μm, 300 s), respectively. At the amplitude of 8 μm an increase in COD value in leachate was the lowest and the time of ultrasonication was the longest (420 s).

Figure 3 shows the dependence of the parameters of ultrasonication during anaerobic process on removal efficiency of pollutants from sonicated wastewater.

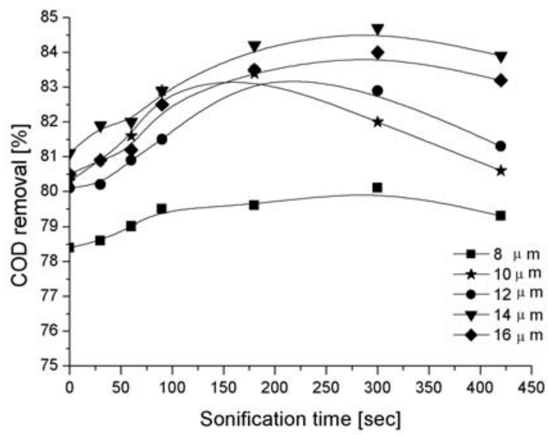

Fig. 3. Ultrasonication time and amplitude versus COD removal

It was found that ultrasonication time of 300 s and higher values of amplitude, i.e. 14 μm and 16 μm, were the optimal parameters of ultrasonication of the leachate following anaerobic digestion. In this case, the COD in effluent was 382.2 mg O_2/dm^3 (COD removal of 84.7%) and 399.6 mg O_2/dm^3 (COD removal of 84.0%), respectively. In the leachate that was not subjected to ultrasonication, the efficiency of COD removal reached 80% (COD value in effluent, 499.6 mg O_2/dm^3).

In the next step of our investigation, anaerobic digestion at optimal parameters of ultrasonic field was studied. Synthetic wastewater containing 20% (v/v) of non-ultrasonicated leachate was digested in four flask. In another four flasks, anaerobic digestion was undertaken under the same conditions, but leachate before biochemical stabilization was subjected to ultrasonication for 300 s at the amplitude of 14 μm. The results are shown in tables 2 and 3.

Table 2

Parameters of anaerobic digestion of ultrasonicated leachate

Parameter	Ultrasonicated landfill leachate before fermentation	Ultrasonicated landfill leachate after fermentation			
		Fermentation time 2 [d]	Fermentation time 4 [d]	Fermentation time 6 [d]	Fermentation time 8 [d]
pH	8.3	7.9	7.9	8.5	8.1
Alkalinity [mg $CaCO_3$ dm^{-3}]	2350	2230	2300	2250	2250
VFA [mg CH_3COOH dm^{-3}]	359.5	531.4	357.5	334.3	321.4
COD [mg O_2 dm^{-3}]	2522.5	831.0	601.2	431.5	301.1
$N–NH_4^+$ [mg dm^{-3}]	275.9	–	–	–	265.7
Chloride [mg dm^{-3}]	582.0	586.0	580.0	590.0	600.0
Biogas [dm^3]	–	1.0	0.09	0.06	0.01

It was proved that ultrasonic disintegration of leachate has positive impact on the efficiency of anaerobic digestion. Hydrolysis of leachate was enhanced, which resulted in a better and faster degradation of organic compounds (figure 4). The highest organic compounds (COD) and VFA concentration and biogas generation were measured on the second day of digestion of the leachate both ultrasonicated and non-ultrasonicated. However, in anaerobic digestion of non-conditioned leachate, a decrease in COD was lower, i.e. 81.1%. In the case of leachte subjected to ultrasonification, COD removal was higher, i.e. 88%.

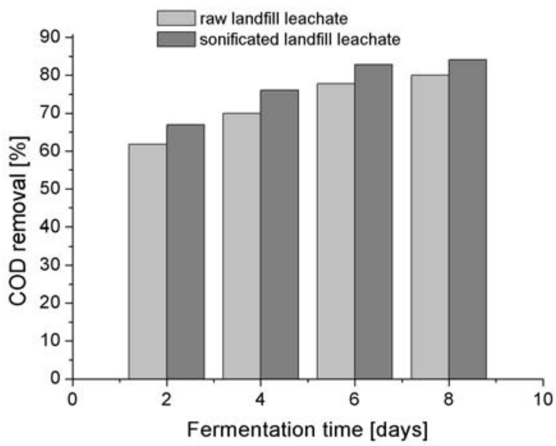

Fig. 4. Comparison of COD removal efficiency during fermentation process of raw and ultrasonicated landfill leachate

Finally also a better efficiency during acitogenesis of the ultrasonicated leachate was observed. In that case, after 8 days of fermentation, the value of COD in effluent was 301.2 mg O_2/dm^3, hence COD removal reached 88.0%. In the leachate non-conditioned, COD removal was 462.5 mg O_2/dm^3 (81.0% COD removal). That fact confirmed a positive influence of ultrasounds on hydrolysis phase.

However, in both experiments, reduction of VFA concentration after the fourth day of fermentation was observed. This can be explained by biodegradation of organic compounds being more susceptible to this process.

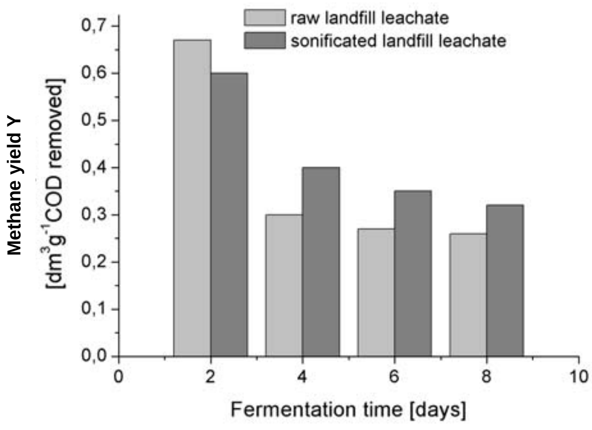

Fig. 5. Methane yield versus fermentation time for raw and ultrasonicated landfill leachate

During all experiments the VFA/alkalinity ratio, which properly represents fermentation, was estimated. The maximum value above which the process inhibition takes place is assumed on the level of 0.3 [16]. The highest value of VFA/alkalinity ratio (0.24) was reached on the second day of experiment, then it was maintained on a constant level in the range of 0.13–0.14 for ultrasonicated leachate and in the range of 0.13–0.18 for non-conditioned wastewater. Neither pH nor alkalinity changed during fermentation.

On the second day of fermentation the biogas production was the highest and on the same level for both processes (1 dm^3). In the second period of the experiment, the biogas production decreased. A total volume of biogas produced during fermentation was 1.13 dm^3 for non-conditioned leachate and 16 dm^3 for ultrasonicated leachate. Figure 5 presents specific methane yield calculated for the experiment conducted.

Table 3

Anaerobic digestion parameters of raw leachate

Parameter	Raw landfill leachate before fermentation	Raw landfill leachate after fermentation process			
		Fermentation time 2 [d]	Fermentation time 4 [d]	Fermentation time 6 [d]	Fermentation time 8 [d]
pH	8.3	7.8	7.8	8.3	8.1
Alkalinity [mg $CaCO_3$ dm^{-3}]	2350	2240	2400	2350	2250
VFA [mg CH_3COOH dm^{-3}]	300.0	415.5	325.7	368.8	368.5
COD [mg O_2 dm^{-3}]	2440.5	930.6	726.2	566.2	462.5
$N–NH_4^+$ [mg dm^{-3}]	261.8	–	–	–	260.4
Chloride [mg dm^{-3}]	580.0	–	–	594.0	595.0
Biogas [dm^3]	–	1.0	0.06	0.05	0.02

Methane yield of conditioned leachate was by about 22% higher compared to that of non-conditioned.

4. SUMMARY

It is known that the length of fermentation period, the value of the specific methane yield and the efficiency of organic compounds' removal strongly depend on leachate biodegradability. The results obtained during our experiment reveal that ultrasonic disintegration of landfill leachate improves the biodegradation efficiency of organic compounds. It can be assumed that ultrasonication changes chemical structure of slow biodegradating compounds, hence their anaerobic degradation is more efficient.

It was proved that 300 s ultrasonication of leachate at the amplitude of 14 μm increased the COD removal efficiency by 7% compared with the fermentation of non-conditioned wastewater. In that case, the efficiency of COD removal reached 88.0% and the concentration of organic compounds in effluent was 301.2 mg O_2/dm^3. An increase in the biogas production (2.6%) was achieved on the 8th day of the process when specific methane yield in ultrasonicated leachate was by 22% higher compared with that in non-conditioned leachate.

Improvement in the efficiency of anaerobic digestion by optimizing the fermentation parameters is possible and it will be studied.

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