Hindawi Publishing Corporation
Journal of Chemistry
Volume 2014, Article ID 483946, 1 page
http://dx.doi.org/10.1155/2014/483946

Retraction

Retracted: Improvement of Landfill Leachate Biodegradability with Ultrasonic Process

Journal of Chemistry

Received 5 March 2014; Accepted 6 March 2014; Published 31 March 2014

The paper titled "Improvement of landfill leachate biodegradability with ultrasonic process" has been retracted as it is essentially identical in title and technical content with a previously published paper "Mahvi AH, Roodbari AA, Nabizadeh Nodehi R, Nasseri S, and Dehghani MH et al. (2012), Improvement of landfill leachate biodegradability with ultrasonic process, PLoS ONE 7(7): e27571.doi: 10.1371/journal.pone.0027571," [1] and it was found of significant similarity with the published article "A. Roodbari, R. Nabizadeh Nodehi, A. H. Mahvi, S. Nasseri, M. H. Dehghani, and M. Alimohammadi, Use of a sonocatalytic process to improve the biodegradability of landfill leachate," Braz. J. Chem. Eng. vol. 29 no. 2 São Paulo Apr./June 2012, http://dx.doi.org/10.1590/S0104-66322012000200003.

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[1] M. Amirhossein, R. Aliakbar, N. N. Ramin, N. Simin, D. Mohammadhadii, and A. Mahmood, "Improvement of landfill leachate biodegradability with ultrasonic process," *E-Journal of Chemistry*, vol. 9, no. 2, pp. 766–771, 2012.

http://www.ejchem.net

ISSN: 0973-4945; CODEN ECJHAO
E-Journal of Chemistry
2012, **9(2)**, 766-771

Improvement of Landfill Leachate Biodegradability with Ultrasonic Process

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Received 23 August 2011; Accepted 4 October 2011

Abstract: Leachate from mature landfills is typically characterized by high ammonium (NH_4^+) content, low biodegradability (low BOD_5/COD ratio) and high fraction of refractory and large organic molecules such as humic and fulvic acids. Mature leachate effluents are known to contain recalcitrant and/or non-biodegradable organic substances and biological processes are not efficient in these cases. A promising alternative to complete oxidation of biorecalcitrant leachate is the use of ultrasonic process as pre-treatment to convert initially biorecalcitrant compounds to more readily biodegradable intermediates, followed by biological oxidation of these intermediates to biomass and water. The objectives of this study are to investigate the effect of son catalysts process on biodegradability improvement. Results showed that when applied as relatively brief pre-treatment systems, the sonocatalysis processes induce several modifications of the matrix, which results in significant enhancement of its biodegradability. For this reason, the integrated chemical–biological systems proposed here represent a suitable solution for the treatment of landfill leachate samples with an efficient remediation of the relevant parameters (COD, TOC).

Keywords: Biodegradability, Improve, Leachate, Ultrasonic.

Introduction

The generation of leachate remains an inevitable consequence of the practice of waste disposal in sanitary landfills[1,2]. Biological treatment of leachate is often the most cost-effective alternative when compared to other treatment options. Traditionally, the degradation of organic compounds and the removal of nitrogen can be achieved by advanced oxidation processes (AOP)[3,4]. Ultrasonic process is one of AOP and involve the generation of the hydroxyl radical (•OH) and pirolysis phenomenon, which has a very high oxidation potential and is able to oxidize almost all organic pollutants and volatile matter such as NH_3. Although these processes are very effective in completing mineralization of pollutants, if they are applied as the only treatment process, they will be expensive[5]. A promising alternative to

complete oxidation of biorecalcitrant leachate is the use of ultrasonic process as pre-treatment to convert initially biorecalcitrant compounds to more readily biodegradable intermediates, followed by biological oxidation of these intermediates to biomass and water[6,7]. Biological treatment methods used for the leachate treatment can be classified as aerobic, anaerobic and anoxic processes which are widely used for the removal of biodegradable compounds[8]. Physicochemical methods are used along with the biological methods mainly to remove non-biodegradable compounds from the leachate[9,10,11]. As a result, parameters have been developed to characterize leachate and predict its treatment efficiency. The ratio of biochemical oxygen demand (BOD) to chemical oxygen demand (COD; BOD/COD) is a common classification approach. Leachate is classified as stabilized, intermediate, or fresh given BOD/COD values of <0.1, 0.1–0.5 and >0.5, respectively[12,13,14]. The objectives of this study are to investigate the effect of ultrasonic process on leachate biodegradability improvement.

Experimental

Samples of landfill leachate were obtained from a municipal landfill site located in Shahrood (Semnan, Iran). All leachate samples were collected from leachate lift stations or storage tanks, stored at 3 °C and tested within 2 d of collecting the samples. The ammonia- nitrogen concentrations were analyzed with C_{203} 8 parameter test meter (Hanna electronics co., Ltd.). The pH was measured by a Benchtop pH Meters (Cole-Parmer Co., Ltd).The pH meter was calibrated before each use with pH 3, 7 and 10 buffer solutions. BOD and COD measurements were determined following Standard Methods 5210 and 5220, respectively[14].

Experimental set-up

As shown in Figure 1, for the laboratory experiments a cylindrical shape Plexiglas reactor with total volume of 1 L was prepared. The solution in the reactor was mixed with a magnetic stirrer, while sufficient aeration was provided by a compressor connected to a porous stone located in the bottom of the reactor. The compressor was used to ensure completely mixed condition in the reactor. The ultrasonic source was a Model UGMA-5000 ultrasound generator with three 30, 45 and 60 kHz transducers having a titanium probe with 20 mm diameter. The power input could be adjusted continuously from 60 to 120 W. A leachate sample of 1000 mL was sonicated in a covered cylindrical glass vessel. Aeration was supplied by a Model SALWAT air compressor.

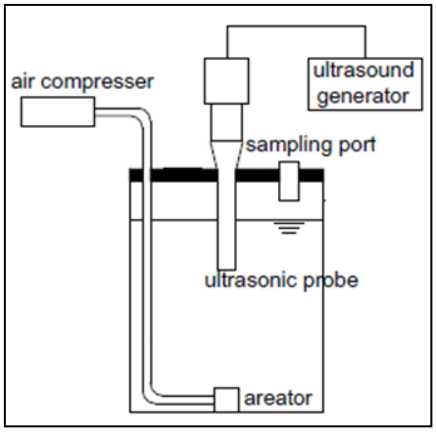

Figure 1. Schematic diagram of the reactor.

Procedure

After the optimization by factorial design, the ultrasonic were applied in the treatment of raw leachates using a batch wise mode. At first, the raw leachate sample was filtered by filter paper (0.45 μ) to remove any suspended solid impurity[15]. Then the sample was adjusted to the required pH with H_2SO_4 or NaOH. Then different scenarios were tested with regard to power intensities of 70 and 110 W, frequencies of 30, 45 and 60 KHz, reaction times of 30, 60, 90 and 120 minutes and pH of 3, 7 and 10. For determining the effects of catalysts on sonication efficiencies, 5 mg /L of TiO_2 and ZnO have been also used.

Results and Discussion

Characterization of the raw landfill leachate

Main chemical characteristics of raw leachate summarized in Table 1. With biodegradability ratio (BOD5/COD) lower than 0.35 and a pH higher than 8, the samples can be considered as moderately stabilized leachates[16,17], normally classified as refractory to conventional biodegradation processes.

Table 1. Chemical characteristics of the studied landfill leachates.

Parameters	Values	Parameters	Values
COD	5691±83	pH	7.9-8.1
Calcium	10.61±0.2	Magnesium	8.65
BOD_5	1738±36	NH3-N	726±25
TOC	1536±20	TS	1420±29
Alkalinity as CaCO3	3650±123		

Values (except pH) in mg /L.

Effect of sonocatalyst on biodegradability of leachate

The results indicated that sonocatalyst process can improve leachate biodegradability (BOD_5/COD ratio). BOD_5/COD ratio for raw leachate was 0.35 but it reached to 0.786 (with TiO_2) and 0.783 (with ZnO) after sonication. Independent Samples T-test showed there is significant difference between BOD_5/COD ratio of raw leachate and pretreated leachate with sonocatalysis process. (p_{value}=0.000 for both TiO_2 and ZnO).

Effect of ultrasound power input on biodegradability improvement

As shown in Figure 2, the power input clearly improves biodegradability. Independent Samples *T*-test showed there is significant difference between BOD_5/COD ratio of raw leachate and pretreated leachate at different powers. (p_{value}=0.000 for both TiO_2 and ZnO). According to sonochemistry theory, when the ultrasound intensity reaches or exceeds the cavity threshold, bubbles will be formed easily and the cavities collapse violently[18]. Increasing the ultrasonic power will increase the energy of cavitation, lowering the threshold limit of cavitation, and enhancing the quantity of the cavitation bubbles[19,20].

Effect of exposure time on biodegradability improvement

As shown in Figure 3, the exposure time improve biodegradability somehow. One-Way ANOVA test showed there is no significant difference between BOD_5/COD ratio of raw leachate and pretreated leachate with sonocatalysis process at different exposure times. (p_{value}=0.467 for TiO_2 and 0.398 for ZnO).

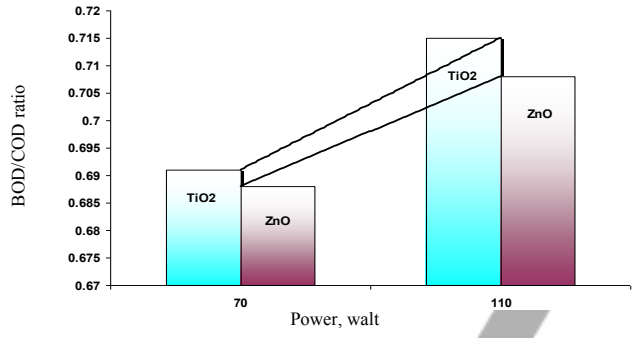

Figure 2. BOD/COD ratio input for TiO_2 and ZnO at different powers (Frequency=30KHz, concentration= 5 mg/L, pH=3).

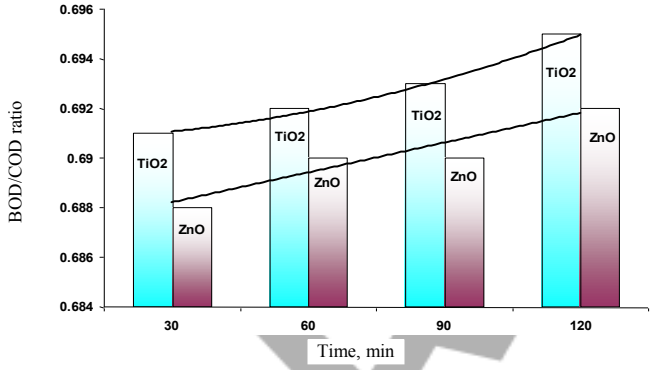

Figure 3. BOD/COD ratio for TiO_2 and ZnO at different exposure times (Frequency= 30Khz, concentration= 5mg/L, pH=3).

Effect of frequency on biodegradability improvement

As shown in Figure 4, the frequency improves biodegradability. One-Way ANOVA test showed there is significant difference between BOD_5/COD ratio of raw leachate and pretreated leachate with sonocatalysis process at different frequency. (p_{value}=0.000 for both TiO_2 and ZnO). Results of Tukey statistical test also showed that there are significant difference between frequencies of 30 and 60 KHz (P_{value}=0.000) and 45 and 60 KHz (P_{value}=0.000).

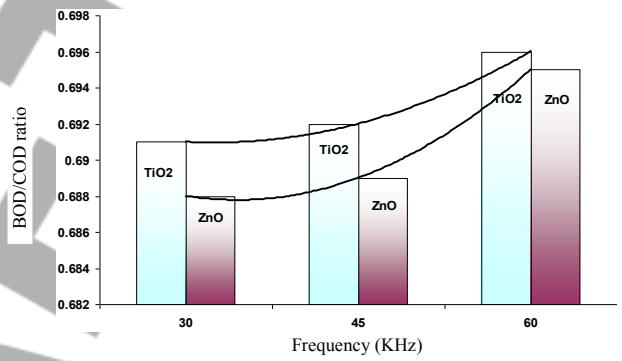

Figure 4. BOD/COD ratio for TiO_2 and ZnO at different frequencies (power=70watt, concentration= 5 mg/L, pH=3).

Effect of pH on biodegradability improvement

As shown in Figure 5, pH improves biodegradability somehow. One-Way ANOVA test showed there is no significant difference between BOD_5/COD ratio of raw leachate and pretreated leachate with sonocatalysis process at different pH. (p_{value}=0.503 for TiO_2 and0.170 for ZnO).

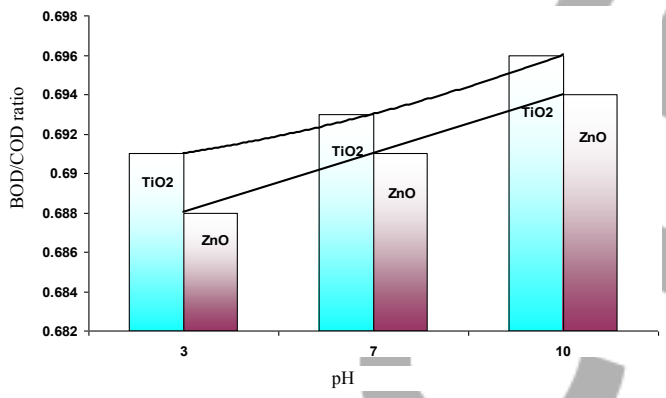

Figure 5. BOD/COD ratio for TiO_2 and ZnO at different pH (power=70watt, concentration= 5 mg/L, frequency=30 KHz).

Effect of type of catalysts on biodegradability improvement by ultrasound

Figure 6 shows the effect of types of catalysts on leachate biodegradability (BOD_5/COD ratio). Results showed that effects of two catalysts on leachate biodegradability was similar but Independent Samples T-test indicated that the there is no significant difference between BOD_5/COD ratio of raw leachate and pretreated leachate with TiO_2 and ZnO. (p_{value}=0.287).

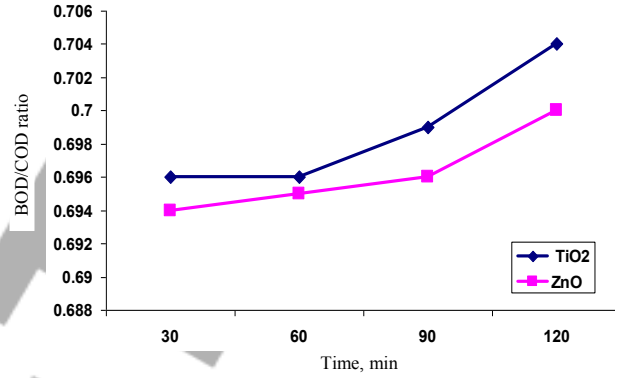

Figure 6. BOD_5/COD ratio for TiO_2 and ZnO (power=70watt, concentration= 5 mg/L, frequency=30 KHz).

Conclusion

Landfill leachates contain some macromolecular organic substances that are resistant to biological degradation. With very low biodegradability ratios (BOD/COD), usually lower than 0.35, these complex matrixes show a recognized resistance toward conventional activated sludge systems. When applied as relatively brief pre-treatment systems, the sonocatalysis processes induce several modifications of the matrix, which results in significant

enhancement of its biodegradability. For this reason, the integrated chemical–biological systems proposed here represent a suitable solution for the treatment of landfill leachate samples with an efficient remediation of the relevant parameters.

Acknowledgment
Authors would like to thank from research affair of Tehran University of medical sciences.

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