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Ultrasound pretreatment for enhanced biogas production from olive mill wastewater

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ABSTRACT

This study investigates applicability of low frequency ultrasound technology to olive mill wastewaters (OMWs) as a pretreatment step prior to anaerobic batch reactors to improve biogas production and methane yield. OMWs originating from three phase processes are characterized with high organic content and complex nature. The treatment of the wastewater is problematic and alternative treatment options should be investigated. In the first part of the study, OMW samples were subjected to ultrasound at a frequency of 20 kHz with applied powers varying between 50 and 100 W under temperature controlled conditions for different time periods in order to determine the most effective sonication conditions. The level of organic matter solubilization at ultrasound experiments was assessed by calculating the ratio of soluble chemical oxygen demand/total chemical oxygen demand (SCOD/TCOD). The results revealed that the optimum ultrasonic condition for diluted OMW is 20 kHz, 0.4 W/mL for 10 min. The application of ultrasound to OMW increased SCOD/TCOD ratio from 0.59 to 0.79. Statistical analysis (Friedman's tests) show that ultrasound was significantly effective on diluted OMW ($p < 0.05$) in terms of SCOD parameter, but not for raw OMW ($p > 0.05$). For raw OMW, this increase has been found to be limited due to high concentration of suspended solids (SS). In the second part of the study, biogas and methane production rates of anaerobic batch reactor fed with the ultrasound pretreated OMW samples were compared with the results of control reactor fed with untreated OMW in order to determine the effect of sonication. A nonparametric statistical procedure, Mann–Whitney *U* test, was used to compare biogas and methane production from anaerobic batch reactors for control and ultrasound pretreated samples. Results showed that application of low frequency ultrasound to OMW significantly improved both biogas and methane production in anaerobic batch reactor fed with the wastewater ($p < 0.05$). Anaerobic batch reactor fed with ultrasound pretreated diluted OMW produced approximately 20% more biogas and methane compared with the untreated one (control reactor). The overall results indicated that low frequency ultrasound pretreatment increased soluble COD in OMW and subsequently biogas production.

1. Introduction

The olive oil industry is one of the important agro-industries in Mediterranean countries including Spain, Italy, Greece, Turkey, Syria and Tunisia. Olive mill wastewater (OMW), which is characterized with high chemical oxygen demand (COD), biochemical oxygen demand (BOD) and suspended solid (SS) concentration as well as a variable amount of lipids and phenolic compounds, is one of the strongest, complex and problematic industrial wastewaters. Several factors such as method of extraction, type and maturity of olives, region of origin, climatic conditions and associated cultivation methods, the technology used for oil extraction have been reported to affect wastewater quality and quantity [1]. The wastewater is produced in large quantities in Mediterranean countries and represents a serious environmental problem if not properly managed. Due to the complex characteristics of the wastewater, the treatment of the OMW is still a major challenge facing olive mill producers. Therefore, OMW is usually collected in central lagoons or stored in small ponds beside the mill where it is left to evaporate during summer season [2]. These ponds cause considerable harmful effects to the environment such as colouring of natural water, threat to aquatic life, causing surface and ground water pollution, changing soil quality/plant growth and causing odours [3].

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Due to increasing environmental constraints, there is a growing interest in the application of new technologies for the treatment of the OMW. Therefore, several treatment options and strategies including physical [4], chemical [5], biological [6,7] and the combinations of the methods [8] have been proposed or tested for management of the wastewater. However, up to now, a viable wastewater treatment scheme could not be proposed for the wastewater in order to meet standards of environmental regulations. All the proposed schemes have several disadvantages. Chemical processes have a disadvantage of both high operating costs and high sludge quantities which requires additional treatment thereby additional investment and high operating cost. Aerobic biological treatment processes can be used for OMW, only if high dilution ratios and/or pretreatment were applied [9]. Another disadvantage is high operating cost both for aeration of the wastewater and treatment of sludge produced in huge amounts during the process.

Among biological processes, anaerobic treatment is a viable option for OMW due to high organic content. Therefore, many studies have been conducted using anaerobic treatment for OMW in order to remove the high organic load efficiently [10–15]. However, there are still some concerns in the application of biological processes to the wastewater due to possible detrimental effect of the compounds present in the wastewaters such as dark coloured polyphenolic compounds [16]. Another problem is high concentration of particulate organic solids in OMW [17] which is not possible to settle the suspended solids in OMW with conventional sedimentation tanks. The particulate organic matter limits hydrolysis stage of anaerobic digestion [18,19]. Both concentration and content of particulate solids are important factors in hydrolysis. Therefore, enhanced performance could be achieved by combining a pretreatment step prior to anaerobic process to increase hydrolysis step. It has been stated that anaerobic filters or up-flow anaerobic sludge bed reactors are suitable systems for the treatment of OMW [2]. However, they also added that integration of a pretreatment stage to the anaerobic processes is necessary. Owing to the complexity of the wastewater, it is urgent to investigate new technologies for the management of the wastewater. Combination of many pretreatment methods was tested for anaerobic processes [20,21] but most of them are unviable. Ultrasonic pretreatment before anaerobic processes is one of the methods that remain unexplored for the OMW.

Ultrasound which is a sound wave at a frequency range from 20 kHz to 10 MHz have a wide range of environmental applications. Low frequency ultrasonic pretreatment prior to anaerobic sludge digestion is one of the most promising recent technologies which have been extensively investigated for wastewater sludge management [22,23]. It has been stated that the most efficient solubilization was achieved by the lowest frequency [22]. It has been shown that ultrasonic pretreatment of both primary sludge [24,25] and secondary sludge [24] enhances the performance of anaerobic digestion by accelerating the hydrolysis step due to the increase in bioavailable substrate concentration [26]. Several researchers reported decreased sludge particle size [24] and soluble COD increase in the supernatant due to low frequency US pretreatment [22]. Beside sludge management, in the last decade, there is a growing interest in application of ultrasound technology for water and industrial wastewater treatment to improve biodegradability and reduce the toxicity of different industrial wastewaters [27–29]. The process can also be used as a pretreatment step to enhance hydrolysis rate of OMW in anaerobic processes. In the literature, there are some studies on the effect of ultrasound on wastewater treatment [30–33]. However, studies on using ultrasound as a pretreatment step in wastewater management prior to anaerobic batch tests are scarcely available [34,35]. Although there are several studies on application of US for OMW [31–33], to our knowledge, the combination of ultrasound with anaerobic batch processes for the wastewater has not been reported yet.

This work, therefore, investigates application of low frequency ultrasound to OMW as pretreatment step in order to enhance the hydrolysis stage in anaerobic batch tests. The potential efficiency of ultrasound on solubilization of organic compounds in OMW has been specifically investigated in terms of COD, soluble COD and suspended solids under different US energy inputs and durations.

2. Materials and methods

2.1. Wastewater source

OMW was obtained from an olive oil extraction plant located near the city of Canakkale, Turkey, which uses a three phase process at November, 2012. The sample was collected as composite during the production process.

2.2. Sources of inoculum sludges

Anaerobic seed sludge was collected from a full-scale Expanded Granular Sludge Bed (EGSB) reactor used in the anaerobic stage of a two stage anaerobic–aerobic biological treatment system at a brewery and used as seed sludge in the anaerobic batch studies. Since the reactor sludge has a granular structure, total solids (TS) and total volatile solids (TVS) were measured for determining solid concentration. TS and TVS concentrations of the anaerobic granular sludge were 37.22 ± 7.28 g/L and 31.33 ± 6.82 g/L, respectively.

2.3. Analytical methods

COD concentrations were measured with a Hach spectrophotometer (Hach Lange DR 5000) and vials for COD between 0–800 ppm. Soluble COD was determined after filtering the samples through 0.45 μm filter paper. pH measurements were taken with a pH meter (ORION SA 520 pH meter). SS (2540 D), TS and TVS concentrations (2540 B) were measured following standard methods [36]. Total organic carbon (TOC) and total nitrogen (TN) concentrations were also determined by TOC–TN equipment. Total phenolic concentrations were quantified by means of Folin–Ciocalteau colorimetric method using gallic acid as standard [37]. Ortho phosphate, phosphate, nitrate parameters were measured using kits (Hach-Lange).

In anaerobic batch reactors, biogas production was continuously measured using MilliGascounter MGC-1 (Ritter, Bochum, Germany). Biogas composition including methane, carbon dioxide, hydrogen and nitrogen was determined by a gas chromatograph (Agilent 7820A) equipped with a thermal conductivity detector (TCD) and a molecular sieve column (HP INNOWAX 30 m × 0.25 mm and 30 m × 0.50 mm).

2.4. Ultrasound

Ultrasound experiments were performed using a generator (Vibra Cell505) combined with a transducer and a metallic probe of 1.9 cm in diameter. This apparatus works with a frequency of 20 kHz and a power of 500 W. The studies were carried out at the average power densities of 0.2 and 0.4 W/mL for 250 mL sample volumes. A water-cooling bath was used for controlling the temperature increase due to ultrasonic heating in the wastewater samples. The temperature of the treated samples did not exceed 35 °C during the experiments. Reactors used in the experiments were all made of dark-colored amber glass with a total volume of 250 mL.