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Evaluating the effect of ultrasonication on sewage biodegradation by measuring particle size distribution

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ABSTRACT

Application of ultrasonication in optimum conditions as a pretreatment step before activated sludge process could increase the readily biodegradable fraction of wastewaters, but optimization of ultrasonication requires a reliable and simple control parameter. There is a direct relationship between biological degradation rate and particle size of organic substrates and distribution of chemical oxygen demand (COD) in terms of particle size in a wastewater can be determined by particle size distribution (PSD) analysis. The objective of this study is to investigate the applicability of PSD as a tool for monitoring the effect of ultrasonication on the biological treatability of municipal wastewaters. A municipal wastewater sample taken from an activated sludge process influent with high particulate COD (75.4% of 690 mg/L total COD) was subjected to ultrasonic pretreatment with 35 kHz frequency and 0.25 W/mL constant specific power for 2, 4, and 8 min reaction periods. After pretreatment, PSD of each ultrasonicated wastewater sample was determined by sequential filtration–ultrafiltration using eight filters with different pore sizes between 700 and 2 nm. The particulate COD sourced from the organic particles above 700 nm decreased gradually with increased ultrasonication periods from 520 to 326 mg/L (47.2% of total COD), while soluble COD (<2 nm particle size) increased from 85 (12.3%) to 142 mg/L (20.6%). The experimental results demonstrated that PSD analysis can reflect the disintegration of organic particles by ultrasonication and there is a linear relationship between sonication period and COD concentration variations of particle size intervals. Therefore, PSD analysis can be used as a reliable tool for determination of optimal ultrasonication conditions.

Keywords: Sewage; Ultrasonication; Particle size distribution; Biodegradability; Disintegration; Hydrolysable COD

1. Introduction

Biological treatment is the most commonly used treatment alternative, especially for municipal wastewaters, because of its numerous advantages. Despite its existing advantages, there are many studies aiming to increase the treatment efficiency, while minimizing the activated sludge process in terms of either tank volume or aeration requirements [1]. Together with technological advances, understanding the characteristics of organic pollutants in wastewater in relation to the metabolisms of micro-organisms is also very important for the development of biological treatment. Therefore, wastewater characterization and especially chemical oxygen demand (COD) as the most useful parameter representing the organic constituents in wastewater are very important components of the studies on the design and optimization of biological treatment. However, total COD cannot identify the bacterial growth limiting components directly related to process rate and hence treatment plant dimension. For this reason, the COD parameter was divided into fractions, in order to represent readily and slowly biodegradable and nonbiodegradable (inert) organic components [2].

On the other hand, there is a direct relationship between biological degradation rate and particle size of organic substrates. While relatively small molecules can be readily

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transported into microbial cells, larger and more complex molecules require extracellular breakdown by hydrolysis to smaller structures prior to uptake and utilize by microorganisms [3]. Therefore, utilization of soluble readily biodegradable COD can be completed in minutes, but hydrolysis of larger particles takes more time and hence the rate-limiting step of the biological treatment is hydrolysis [2]. Acceleration of this rate-limiting step means reducing the volume, aeration capacity, and energy required for aeration of activated sludge.

Ultrasonic treatment can be effectively used to destruct complex organic compounds. In this process, acoustic cavitation is generated by pressure variations in a liquid using sound waves with a frequency in the range of 16 kHz–100 MHz, named as ultrasound [4]. It should be noted that, when the ultrasonication is considered as a standalone treatment process, the required time-scale and amount of energy required for complete mineralization of the pollutants are not economically acceptable [5]. Therefore, ultrasound should be considered either as a pretreatment step or in combination with other treatment methods. First studies on ultrasound process were intended to research of its application on sewage sludge as a pretreatment prior to anaerobic digestion [6–9]. Afterwards, many studies on various industrial wastewaters were carried out in which ultrasound was used as pretreatment or cotreatment process together with biological [5,10–18] or chemical/physicochemical processes [19–24]. Most of these researchers concentrated on to improve biological/chemical treatability or treatment efficiency of industrial wastewaters generally containing higher organic pollution than municipal wastewaters. However, due to high flow rates of big cities, municipal wastewater treatment plants may accept higher organic loads than industrial wastewater treatment plants. Also, hydrolysable fractions of these organic loads could have ratios as high as 80% [2]. This character requires sufficient aeration volume to complete the rate-limiting hydrolysis step. Therefore, activated sludge tanks and aeration capacities of municipal wastewater treatment plants are generally much greater than the ones in industrial wastewater treatment plants.

Ultrasonic pretreatment may be used to reduce aeration tank volume by disintegrating the organic particles and converting them from hydrolysable fraction to readily biodegradable form in municipal wastewaters. But, due to its economical disadvantages, ultrasound pretreatment process should be optimized very well. In the activated sludge systems, optimization of ultrasound process should be done considering the enhancement of biological degradation rate. The most applied method to determine the degradation rate of organics bases on the determination of readily and slowly biodegradable/hydrolysable COD fractions using respirometric tests. But, this is not an easy applicable method with its complexity and requirements such as laboratory instruments and model simulations. Distribution of COD in terms of particle size in a wastewater can be determined by particle size distribution (PSD) analysis. This test obtains the COD concentrations of each particle size interval [25]. Because PSD does not give precise information about biodegradability of organic constituents of wastewater, it is more useful and reliable to characterize a wastewater for biological treatment, when PSD is combined with respirometric measurements [26]. Certainly, the particle size of organic solids is not the only factor affecting its degradation rate; some other properties of organics are also important, such as their nature and molecule structure. Nevertheless, it is possible to evaluate the changings in biological degradation rates practically observing the PSD data for a selected wastewater, because particle size reduction will always affect degradation rate positively, less or more.

Studies on the ultrasound application for pretreatment of municipal wastewaters or its relationship with PSD are very rare and one of them was carried out by Gibson et al. [27]. In this study, they aimed to improve disinfection efficiency by breaking the particles using ultrasound and determined the relationship between ultrasonic energy density and particle size variations. Similarly, Yagci and Akpinar [28] investigated the reduction of particle size by ultrasound pretreatment in waste activated sludge. In both studies, particles were characterized with their number in each size interval. However, the more proper method in terms of biological degradability is PSD analysis in which the distribution of COD on particle size intervals is determined in wastewater samples by sequential filtration/ultrafiltration.

Consequently, the aim of this study is to disintegrate the organic particles in the wastewater by ultrasonic pretreatment for increasing their biological degradation rate and to investigate the applicability of PSD analysis as a practical method for detection of disintegration effects provided by this pretreatment method. In this context, a municipal wastewater was characterized both conventional and PSD based. Then, PSD profiles of the wastewaters pretreated by applying ultrasound in different reaction periods were compared to evaluate the destruction effects of ultrasound.

2. Materials and methods

2.1. Wastewater sample

The experiments were carried out on the wastewater sample taken from Nevsehir municipal wastewater treatment plant, which accepts average 20,000 m^3/d wastewater flow rate sourced from Nevsehir city center with a population of 100,000 and located in Middle Anatolian Region of Turkey. The plant is consisting of screens, aerated grit chamber, extended aeration activated sludge unit, and final settling tanks. The wastewater was taken as a grab sample from the inlet of activated sludge aeration tank and stored in 4°C throughout the experimental studies. Composite sampling was not preferred because the aim of sampling was to get a wastewater with a high particulate organic content required for experimental objectives, rather than to determine the daily average characterization of the wastewater. All of the conventional characterization parameters were measured according to Standard Methods (SM) [29]. The methods applied for measuring the parameters were SM 4500-H^+.B for pH, SM 2510 B for conductivity, SM 2540 D for total suspended solids (TSS), SM 2540 D and E for volatile suspended solids (VSS), and SM 5220 C for COD. pH and conductivity were measured using Hach Lange HQ40d model portable multimeter. Filtered COD was determined after filtration of wastewater through Millipore AP40 glass fiber filter with a pore size of 0.70 µm. All measurements were performed as triplicate at least.

2.2. PSD-based wastewater characterization

Sequencing filtration procedure from the biggest to smallest pore size was applied for PSD analysis on raw and ultrasonicated wastewater samples. In the first stage of sequencing filtration, Millipore AP40 glass fiber filter having 0.70 μm pore size was used by means of vacuum filtration apparatus. In the following stages, filtration of wastewater samples was continued using membrane filters having different pore sizes by means of Millipore Amicon® Stirred Cell filtration apparatus with 400 mL volume under positive pressure. Nitrogen was used as an inert gas for ensuring maximum 5 bar pressure required for filtration. Membrane filters used for sequencing filtration were Millipore Durapore® HV filter with 0.45 μm pore size, Millipore Durapore® GV filter with 0.22 μm pore size and Millipore Ultracell® Regenerated Cellulose (RC) filters with 100, 30, 10, 3, and 1 kDa pore sizes, respectively. Detailed information about filters including the pore sizes in nanometer as approximately equivalent of kDa unit were given in Table 1 [25]. The samples for COD analyses were collected from every filtration stage after wasting the first 10 mL of filtrate. RC membrane filters (between 100 and 1 kDa pore sizes) were reusable for a lot of times, while the filters having 0.7, 0.45, and 0.22 μm pore sizes were for single use. RC membranes and stirred filtration cell were cleaned with deionized water and sterilized using 70% ethanol solution after every sample filtration and membranes were stored in 10% ethanol solution and +4°C. These filters were regenerated for next sample filtration using 0.1 N NaOH solution. The permeate flux change of each RC membrane discs were monitored before each sample filtration.

2.3. Ultrasonication experiments

Ultrasonication studies were carried out by a laboratory scale Bandelin Sonopuls HD2200 ultrasonic homogenizer equipped with TT 13 titan-sonotrode producing ultrasonic waves with 35 kHz frequency. While the specific power applied on the wastewater sample was kept as constant at 0.25 W/mL, different reaction periods were applied as 2, 4, and 8 min on 500 mL wastewater sample for each experiment set. Specific power and reaction periods were selected considering keeping the energy consumption at a minimal level as well as to get recognizable effects of investigated pretreatment method. Temperature variations were monitored by the measurements before and after every sonication.

Table 2
Conventional wastewater characterization

Parameter	Value
pH	7.76
Conductivity (μS/cm)	1,409
TSS (mg/L)	488
VSS (mg/L)	420
VSS/TSS (%)	86
Total COD (mg/L)	690
Filtered COD (particle size < 700 nm) (mg/L)	170
Filtered COD/total COD (%)	24.6

3. Results and discussion

3.1. Wastewater characterization

Experimental results of conventional wastewater characterization are given in Table 2. These values are compatible with treatment plant monitoring data measured by operators and the wastewater sample can be considered as a medium–high strength wastewater with its 690 mg/L total COD and 488 mg/L TSS concentrations [30]. VSS/TSS ratio was 86% and this ratio means that most of the solids in the wastewater have organic structure. Similarly, 75.4% of total COD is in particular form. Therefore, this sample was evaluated as a suitable wastewater for the experiments of ultrasonic pretreatment prior to biological treatment, with its high particular organic matter content. High conductivity value as 1,409 μS/cm reflects the tap water characteristics of the region obtained from groundwater sources.

PSD-based COD fractionation of the raw wastewater is shown by bar diagrams both as cumulative and differential COD in Fig. 1. According to the experimental results obtained for raw wastewater demonstrated that 75.4% of the total COD is consisting of the organic particles above 700 nm, while the COD sourced from the particles below 2 nm, which could be accepted as soluble COD, has a ratio of 12.3%.

Table 1
Specifications of the filters used in sequencing filtration [25]

No		Filter type	Filter material	Pore size	Pore size (nm)
1	Filtration	Millipore AP40	Hydrophilic glass fiber	0.70 μm	700
2		Millipore Durapore® HV	Hydrophilic polyvinylidene fluoride	0.45 μm	450
3		Millipore Durapore® GV		0.22 μm	220
4	Ultrafiltration	Millipore Ultracell® PL-100	Regenerated cellulose	100 kDa	13
5		Millipore Ultracell® PL-30		30 kDa	8
6		Millipore Ultracell® PL-10		10 kDa	5
7		Millipore Ultracell® PL-3		3 kDa	3
8		Millipore Ultracell® PL-1		1 kDa	2

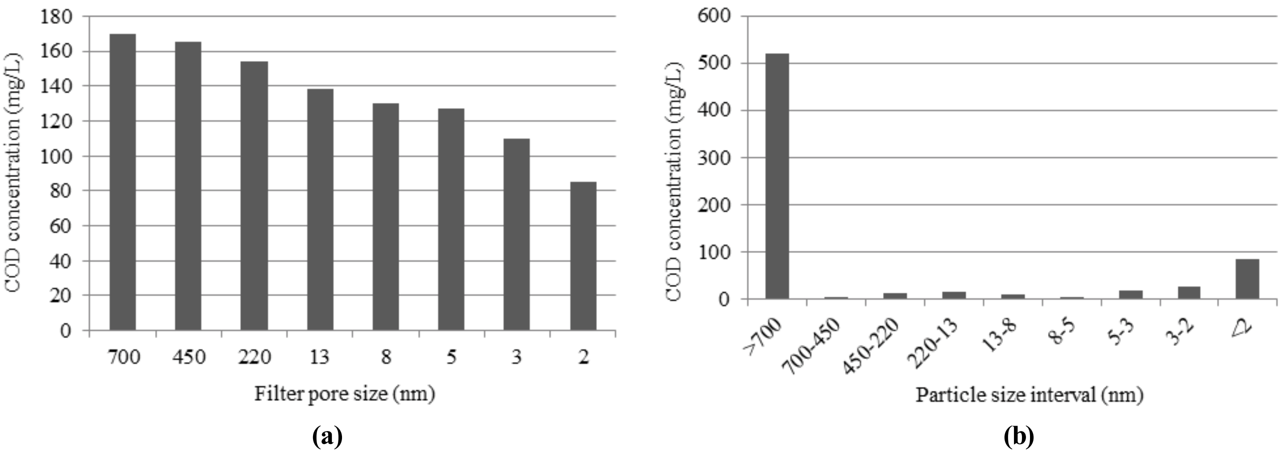

Fig. 1. Particle size-based COD distribution of raw wastewater: (a) cumulative and (b) differential.

The remaining 12.3% of COD dispersed among 2–700 nm particle sizes with lower ratios.

3.2. *Effect of ultrasonication on PSD*

The experimental results obtained from the PSD analyses of raw wastewater and following the ultrasonication experiments with three different reaction times are given in Figs. 2 and 3. The data given in Fig. 2 are the average of the most consistent data selected from the replicates taking into account their standard deviations. Noticeable increase in cumulative COD concentrations can be observed in all particle size intervals as shown in Fig. 2. However, according to Fig. 3, the most apparent effect of ultrasonication occurred in the particles above 700 nm; between raw wastewater and 8 min sonicated wastewater, there is a considerable decrease corresponding to 28.2% in COD concentration from 520 (75.4%) to 326 mg/L (47.2%), respectively. So, it means that 37% of 520 mg/L COD sourced from >700 nm particles in raw wastewater was disintegrated through 8 min sonication and those dispersed to the lower particle sizes. The other particle size intervals which have remarkable changings are 700–450 nm, 220–13 nm, and <2 nm with 7.1%, 8.1%, and 8.3% increasing ratios, respectively. In the other particle size intervals, the changings are in relatively lower ratios.

In Fig. 4, the evolution of COD concentrations with different sonication periods are shown for the particle sizes above 700 nm and below 2 nm which are reflecting particulate and soluble COD fractions and so, the most important particle sizes in terms of biological degradation rate. It can be seen that COD of the particles above 700 nm is decreasing linearly by increasing sonication periods in accordance with first degree reaction kinetic, while COD of the particles below 2 nm is increasing as well. Therefore, this situation demonstrates that ultrasonication is disintegrating organic particles proportional to reaction time and it is possible to detect this

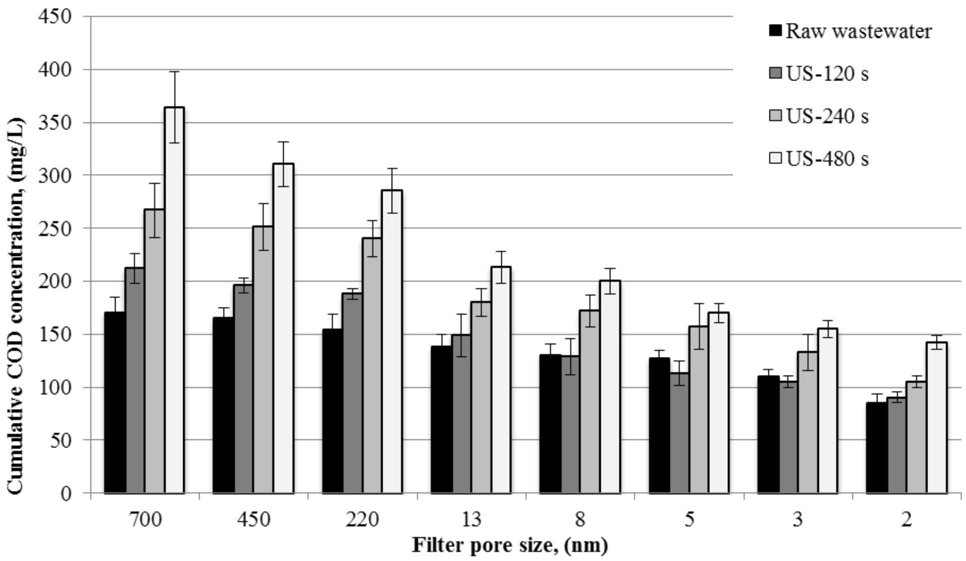

Fig. 2. Cumulative COD distribution of raw and ultrasonicated wastewaters.

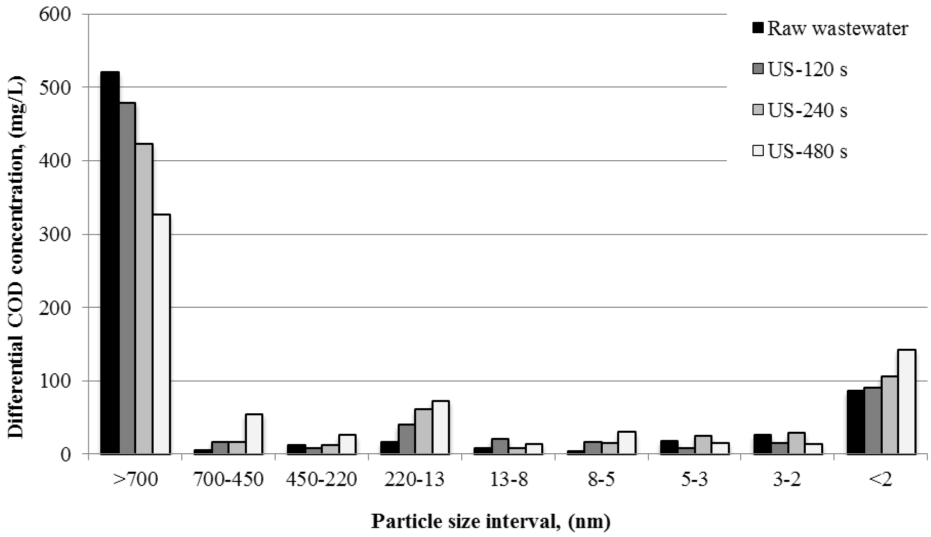

Fig. 3. Differential COD distribution of raw and ultrasonicated wastewaters.

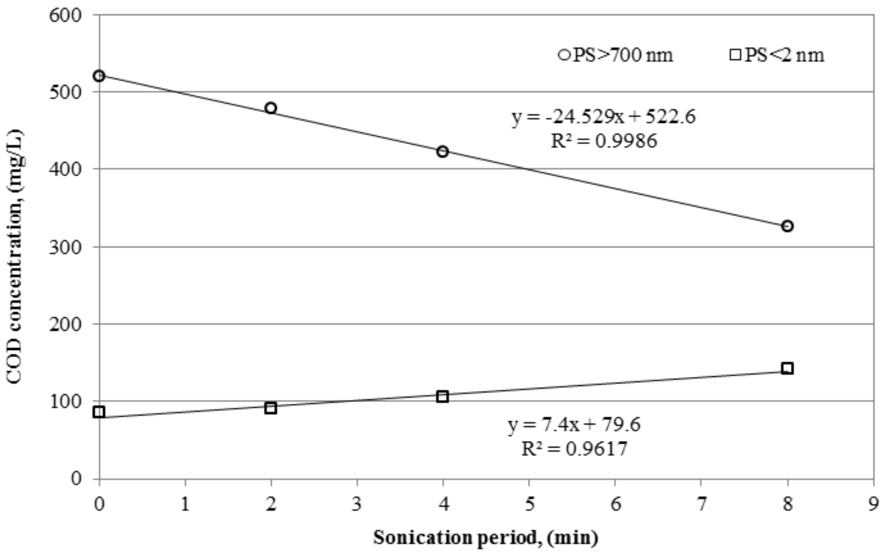

Fig. 4. Evolution of COD concentration with different sonication periods.

effect using PSD analysis. Consequently, PSD analysis could be used as a tool for optimization of ultrasound pretreatment of municipal wastewater.

4. Conclusion

In this study, PSD analysis was used to observe the disintegration effects of ultrasonic pretreatment on organic particles. According to the experimental results, ultrasonic pretreatment can disintegrate the particulate organics in considerable ratios for defined experimental conditions and COD concentration of >700 nm particles decreased to 28.2% by 8 min sonication while soluble COD (<2 nm) concentration increased to 8.3%. There is a linear relationship between sonication period and COD concentrations of particle size intervals and it fits first degree reaction kinetic. This situation provides us to evaluate the effects of ultrasonication on particulate organic matter in a municipal wastewater. Consequently, PSD analysis can be used as a reliable tool for evaluation and optimization of ultrasonic pretreatment of wastewater prior to biological treatment.

The scope of experimental study was quite limited because it was considered as a preliminary study just to demonstrate experimentally the relationship between ultrasonic disintegration and PSD. In the future studies, it is planned to detect the effects of more variables and to support the results in terms of biological degradation rates by respirometric tests, and also to make the cost analysis of ultrasonication pretreatment and hydrolysis process performed biologically to reveal the economic dimension.

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