The Effect of Ultrasound on Particulate Matter, Especially Micro-organisms in Complex Water and Waste Water Media

U. Neis, T. Blume

Technical University Hamburg-Harburg,
Department of Sanitary and Environmental Engineering
21071 Hamburg, Germany
E-mail: neis@tuhh.de

Abstract. Surface waters to be used as resource for drinking water, waste water and sludge produced in treatment processes are very complex media containing soluble, colloidal and particulate matter. Apart from inert or dead matter the colloidal and particulate phase is mainly composed by organic bio-particles (viruses, bacteria, protozoa). Treatment processes like disinfection of potable water, hygienisation of sewage treatment plant effluents or anaerobic sludge stabilisation aim at reducing or inactivating bio-particles. Power ultrasound is used to intensify these processes. Research was conducted to quantify the effects of power ultrasound frequency, intensity and dose. Based on these results the ultrasound reactor design was modified but still has to be optimised.

PROCESS ANALYSIS

When it comes to transforming particulate matter in liquid media by physical, chemical and/or biological reaction processes we observe that transforming particulate matter is usually a rather slow process. It is often the rate determining step with respect to the velocity of the overall removal of contaminants in water or waste water. In this paper we will discuss processes which deal with the treatment of particulate matter, hence we focus on waste water and bio-solids treatment because here the nature of the particles is more diverse and the concentration is much higher. However, the results of our research work might also be applicable for water purification processes like disinfection. Typical data of the composition of untreated municipal waste water is given in Table 1. The table indicates the broad range of inorganic and organic (volatile), of dissolved and undissolved (suspended, settleable) substances normally present in waste water. Normally waste waters are briefly characterised by global parameters like TOC (Total organic carbon), COD (Chemical oxygen demand), BOD (Biochemical oxygen demand), TSS (Total suspended solids), VSS (Volatile suspended solids). dissolved gases like SO_2, CO_2, H_2S, NH_3, O_2, N_2 which, of course, may impact on the cavitation process are not included in this table. We know for example that nitrate/nitrite is formed when water containing molecular oxygen and nitrogen is sonicated [2]. Both substances, nitrate/nitrite, are unwanted in water bodies.

	Concentration (mg/l)		
Constituent	**Strong**	**Medium**	**Weak**
Solids, total:	1200	720	350
Dissolved, total	850	500	250
Fixed	525	300	145
Volatile	325	200	105
Suspended, total	350	220	100
Fixed	75	55	20
Volatile	275	165	80
Settable solids (ml/l)	20	10	5
BOD_5, 20°C	400	220	110
TOC	290	160	80
COD	1000	500	250
Nitrogen, total	85	40	20
Organic	35	15	80
Free ammonia	50	25	12
Nitrites	0	0	0
Nitrates	0	0	0
Phosphorus, total	15	8	4
Organic	5	3	1
Inorganic	10	5	3
Chlorides	100	50	30
Alkalinity	200	100	50
Grease	150	100	50

TABLE 1. Typical composition of untreated domestic waste water [1]

For the sake of convenience the distinction between dissolved and particulate matter is made by filtration of samples through membrane filters with a pore size of 0.45 µm. Of course there are fluctuations of flow and composition of waste water with respect to time and climate (dry weather, rain fall) which impact on the performance of treatment processes. Fig. 1 shows the volumetric particle size distribution in the effluent of the municipal waste water treatment plant in Bad Bramstedt, Germany. Suspended solids are more or less larger than 10 µm, the major part even larger than 100 µm in diameter and most of it is of organic nature (bacteria, protozoa, algae). While single micro-organisms have a size of about 1 to 10 µm, we can assume that the bio-mass in the effluent is present mainly in form of aggregates/flocs.

The elimination of particulate organic matter (pathogens!) is one of the major aims of water treatment. If the removal of such particles is insufficient, they may accumulate in water distribution systems. A number of water purification processes aim at removing or inactivating particulate organic matter like filtration or disinfection. Up to now the application of ultrasound for such kind of water/waste water treatment is rather new. Generally the applied techniques are either of chemical or biological nature. We think that ultrasound offers a great potential for improving water, waste water and sludge treatment processes. First ultrasound applications at technical scale are operational.

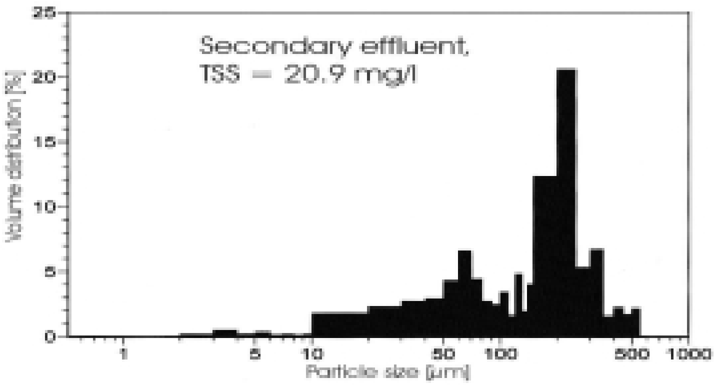

FIGURE 1. Particle size distribution (composite grab sample) of Bad Bramstedt waste water treatment plant effluent.

In Table 2 an overview is given on current ultrasound applications in water, waste water and sludge systems.

Domain	Objective
Potable water	Inactivate bacteria (disinfection)
	Improve separation of solids
	Improve filter regeneration
	Remove incrustations in pipes and wells
Waste water	Sonochemical pollutant degradation
	Improve biological degradation
Biomass/Sludge	Disintegrate bio-solids
	Decompose bulking activated sludge flocs
	Improve dewatering

TABLE 2. Ultrasound applications in environmental engineering

DISINTEGRATION OF PARTICULATE ORGANIC MATTER

Qualitative disintegration model

The production of sewage sludge is a result of mechanical, biological and/or chemical waste water treatment. Sewage sludge is a concentrated suspension of bacterial cells sticking together in a flocculent structure due to extra-cellular polymer substances. If sewage sludge is exposed to ultrasonic energy the first effect is a de-agglomeration of the sludge flocs. The flocs are separated and dispersed without disrupting the cells. After longer treatment time or higher ultrasonic energies, the micro-organism's cell walls are broken and intracellular material is released into the liquid phase.

Consequently, the amount of dissolved organic material is significantly increased [3]. This two-phase model of the ultrasonic disintegration process is illustrated in Figure 2.

The separation of sludge flocs causes a decrease in the average particle size. Assessment of this de-agglomeration process can be done by particle size distribution analysis. If bacterial counts are measured, an increase in counts would be observed when flocs are de-agglomerated into singe cells. Then after longer sonication times the number of bacterial counts would be reduced because the cells are destroyed and the micro-organisms are killed. As mentioned before the size of a single bacteria/micro-organism may lie between 0.5 and 10 µm.

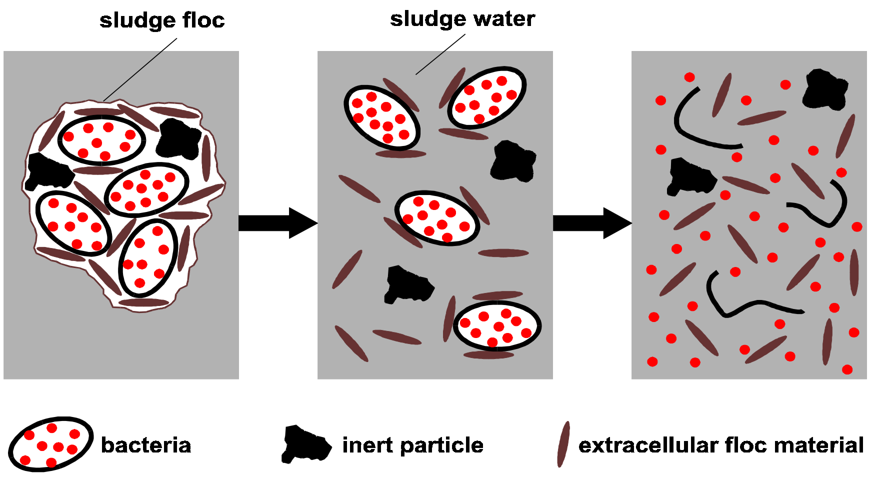

Figure 2. Model of sludge disintegration with increasing ultrasonic specific energy

The extend of cell break-up can be described by several chemical and biological parameters. The cell disruption leads to the release of intracellular organic compounds into the sludge water phase. Therefore, the extend of cell disintegration is determined by the increase of the Chemical Oxygen Demand (COD) in the sludge supernatant. If ultrasonic parameters are chosen appropriately, up to 90% of the total sludge COD could be solubilised [4].
Different sludges contain various types of micro-organisms with different strengths of their cell walls. For the purpose of comparison, a chemical hydrolysis process by incubation of sludge samples in 0.5 mol/l sodium hydroxide for 22 hat 20°C may be done [5]. By this method the effects of cell disruption becomes comparable for different suspensions. A factor DD_{COD}, “degree of disintegration”, is defined as the ratio of COD-increase in the supernatant by sonication or other disintegration techniques related to the COD-increase by the chemical hydrolysis method described above. Other authors defined a solubilisation ratio as concentration of Total Organic Carbon (TOC) in the supernatant after centrifuging the sludge sample divided by the TOC concentration of the original [6].

Impact of ultrasound frequency

We applied ultrasound with varying frequencies in a range from 41 kHz to 3,217 kHz and studied its impact on the sludge disintegration in a lab URS 1000 reactor. As usual the bio-mass concentration of the sludge was high (about 20 g/L). As shown in Figure 3, the degree of bio-solid disintegration (DD_{COD}) decreases with increasing ultrasound frequency: The highest degree of disintegration 80% was obtained at the lowest frequency 41 kHz. It is well known that the resonant cavitation bubble radius increases with decreasing ultrasonic frequency. Upon bubble collapse, hard shock waves are induced by jet streams responsible for the mechanical break-up of the microbiological cell walls. The energy released by a jet stream is a function of the bubble size at the moment of collapse. Certainly the bubble size plays a role when it comes to assess the impact of collapsing cavitation bubbles on micro-organisms. But also the cell size as well as the rigidity of the cell walls and hence the resistance to physical break-up will be important. In that context it is interesting to compare our results which relate to highly concentrated bio-mass suspensions with those published by Clasen [7] who investigated the impact of ultrasound on zoo-plankton organisms which are much larger (between 50 µm and several mm), see Fig. 4.

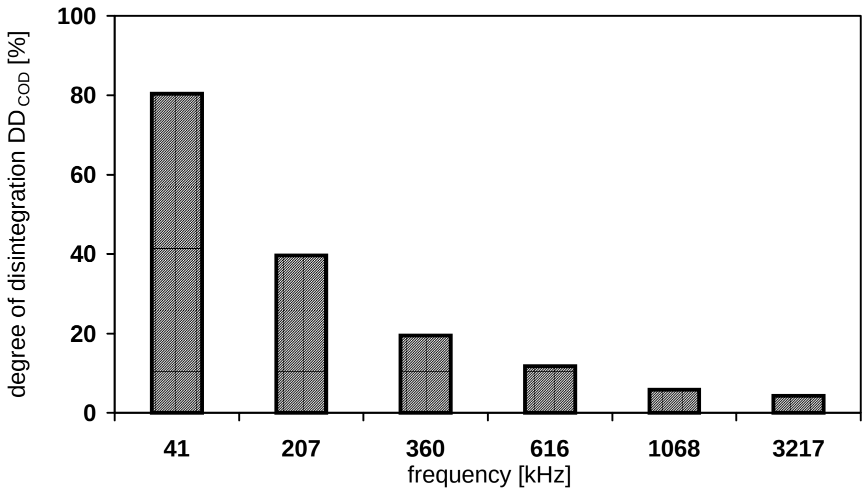

Figure 3. Effect of ultrasonic frequency on the degree of cell disintegration (TSS: 20 g/L, ultrasound intensity: 1.8 W/cm^2, sonication time: 4 h).

The concentration of organisms here is much lower (about 1/1000) as compared to the experiments with sludge bio-mass. All members of zoo-plankton are motile, they are present in surface waters and may disturb the water reclamation process. The active motion enables such organisms to get rid of the fixation in flocs. As it is very difficult to breed the naturally occurring zoo-plankton organisms in great numbers, Clasen used nauplii of the brine shrimp (*Artemia salina*) as model organisms. These can be bred from eggs which are readily available in the pet fish business. Figure 5 shows also that the survival rate of zoo-plankton organisms decreases significantly when the ultrasound frequency is lowered from 100 down to 40 kHz. Obviously the zoo-plankton bodies are damaged in such way that they cannot survive. Unfortunately Clasen does not provide relevant particle size or other detailed analysis so we do not know exactly know how and to what extend cells are torn apart or broken.

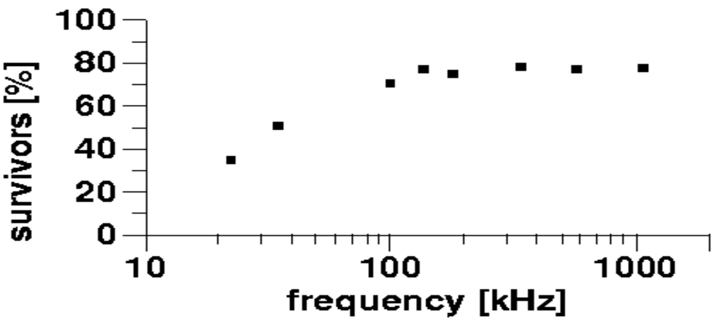

Figure 4. Inactivation of *Artemia nauplii* by ultrasound depending on ultrasound frequency [7].

Impact of ultrasound intensity and dose

We investigated the effect of ultrasound intensity on the disintegration of waste activated sludge and used a 20 kHz laboratory ultrasonic horn (Branson 450, Schwäbisch-Gmünd, Germany). High degrees of disintegration can be obtained by increasing the energy dose. Fig. 5 shows how the degree of disintegration DD_{COD} of waste activated sludge is increased with increasing specific energy input. Within the applied range the intensity did not influence the results when the frequency used is 41 kHz. In this graph disintegration is already observed at a rather low intensity of 0.1 W/cm^2. This is well below the cavitation threshold for water which is about 0.4 W/cm^2. Due to the presence of a high number of solids and gas bubbles, which act as cavitation nuclei in a sludge sample, a lower cavitation threshold seems reasonable. Raising the intensity however must not necessarily produce more intense cavitation. Under certain circumstances the contact between the radiating transducer surface and the liquid can get lost. The motion of the transducer and the liquid get out of phase, an effect which is called de-coupling. Furthermore, higher intensities create more and greater bubbles that may coalesce and lead to less pronounced effects. Therefore the energy transfer efficiency can sometimes be smaller at higher intensities [8]. We did not observed such effects in the reported range between 0.1 and 2 W/cm^2.

Cavitation, induced by ultrasound at low frequencies, is an effective means for the disintegration of living organic cells. Consequently we wanted to investigate whether sonication of water and waste water can be considered as a potential disinfection method. Therefore in lab scale experiments we sonicated samples of different effluents from waste water treatment plants at 20 kHz. These samples were treated for 20 seconds at various ultrasound densities.

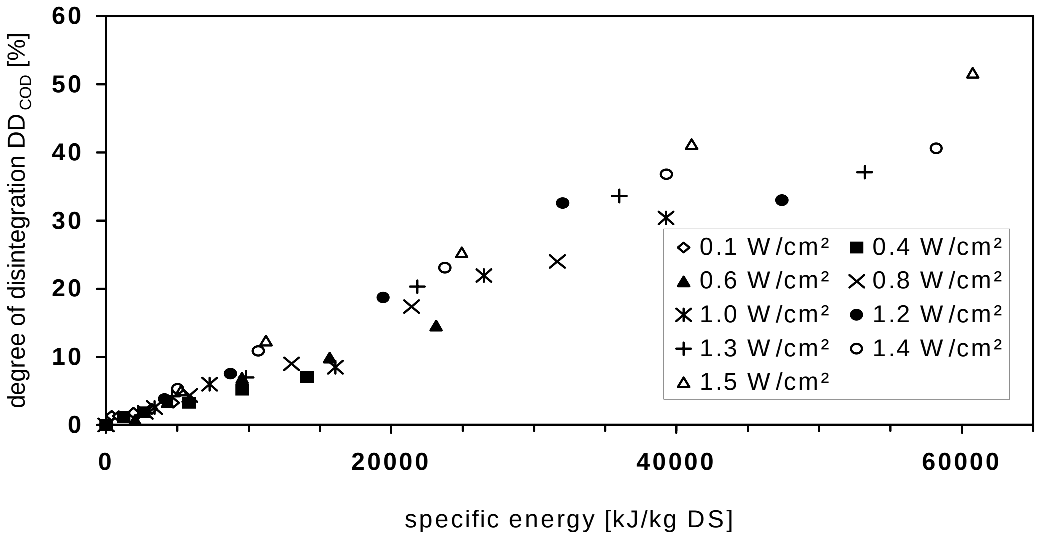

Figure 5. Effect of intensity and energy dose at 41 kHz on the degree of disintegration of sewage sludge bio-solids (lab reactor URS 1000, L-3 Communications ELAC Nautic, Kiel, Germany; sonication time: max. 2 h; bio-solids as dry solids (DS) weight concentration : 25.9 g/kg).

First of all, at low ultrasound intensities a drastic change in samples' particle size distribution was observed indicating efficient de-agglomeration of particle flocs. Initially, 63 % of the solids in the waste water sample were larger than 50 µm in diameter. It stands out that low ultrasound energy (30 W/L) is already sufficient to provoke a clear change in particle composition, whereas further increased ultrasonic doses have only a marginal effect, see Fig. 6. However, all these applied ultrasound doses are too low to have an impact on bacterial counts as they were less than - 0.2 log units (not shown).
For longer sonication times (up to 60 minutes) and maximum US density applied, a significant reduction of microbial counts was observed. Fig. 7 shows that a maximum reduction of 2.9 log units of E.coli was achieved at a dose of 400 Wh/L (60 min at 400 W/L). This is in accordance with the fecal coliforms elimination published by Hua et al. [9].

Fig. 7 shows that at the applied dose fecal streptococci are significantly less vulnerable to cavitation effects than coliform bacteria. This is due to the cell wall structure: gram-positive streptococci's cell walls are notably thicker (20 nm) than gram-negative enterobacteria's (10 – 15 nm), [10]. Several studies have shown that the enterococci species of fecal streptococci are a more appropriate indicator of fecal pollution than fecal coliforms as they show better correlation to human diseases and they survive longer in the water. E. coli as well as fecal streptococci decay kinetics follow a first order reaction behaviour like it is usually observed with other disinfection methods. At the applied ultrasound density of 400 W/L we found the following first order decay rate values k: $k_{E.coli} = 0.11\ min^{-1}$ and $k_{strepto} = 0.03\ min^{-1}$ ($R^2_{E.coli} = 0.96$, $R^2_{strepto} = 0.97$). One might want to compare these results with other data like for UV irradiation:

k = 0.056 min^{-1} at 1 µW/cm² or Ozone: k = 0.88 min^{-1} at 0.5 mg/L, Lezcano et al. [11]. However, it must be considered that these experiments were a first set of tests with lab scale equipment. Experiences with ultrasound sludge disintegration have shown that full scale 5 kW ultrasound reactors are more efficient than lab scale models. Therefore, we hope that ultrasound disinfection efficiency will be better in full scale continuous flow tests.

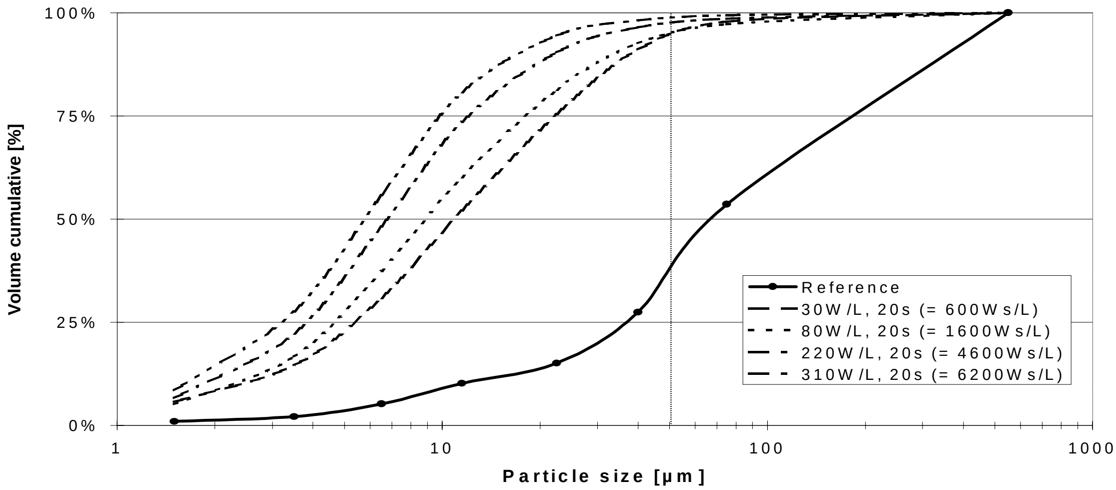

Figure 6. Effect of waste water sonication (20 s at various densities) on Particle Size Distribution

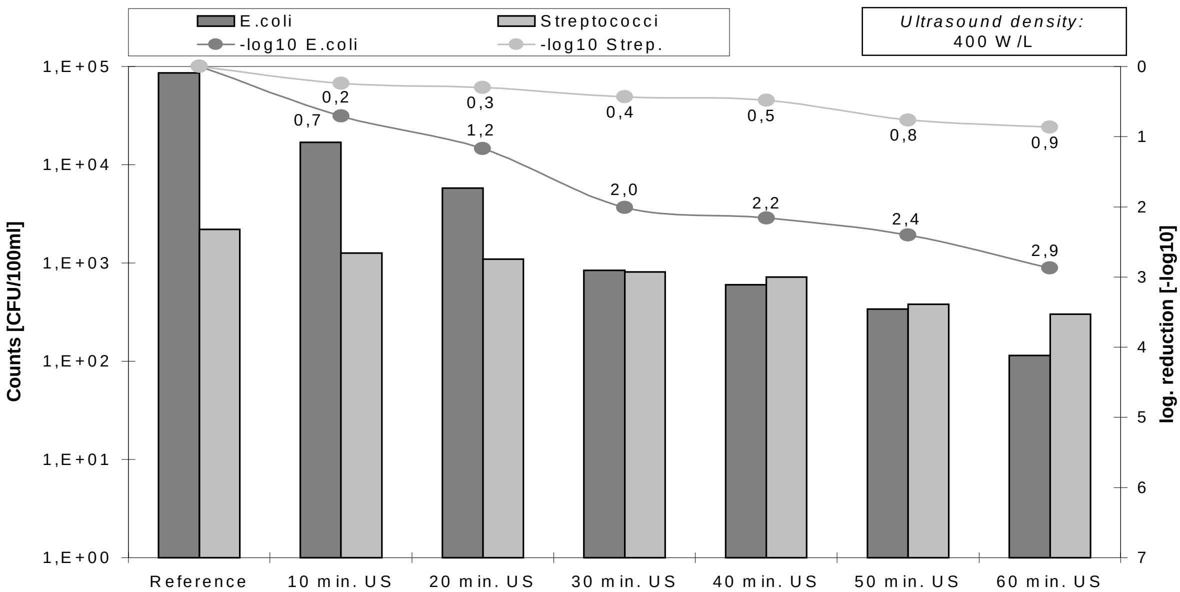

Figure 7. Effect of long sonication on E. coli and fecal streptococci

REFERENCES

1. Metcalf and Eddy, Inc., *Wastewater Engineering: Treatment, Disposal and Reuse* (Tata McGraw-Hill, New Delhi, 1982), ch. 3, 64.
2. Neis U., Tiehm A., (1999) „Ultrasound in waste water and sludge treatement". In: *Ultrasound in Environmental Technology*, ed. by Neis U., Tiehm A., TUHH Reports on Sanitary Engineering**, 25**, Hamburg, ISBN 3-930400-23-5 (1999),. 39-61.
3. Tiehm, A., Nickel, K., Zellhorn, M. and Neis, U. (2001) „Ultrasonic waste activated sludge disintegration for improving anaerobic stabilisation, " Water Research, Vol. 35, **No. 8**, 2003-2009.
4. Clark, C.B. and Nujjoo, I. (1998*) CIWEM Presidential Conference: Treatment innovation for the next century*, Cambridge, UK.
5. Tiehm, A., Nickel, K. and Neis, U. (1997) „The use of ultrasound to accelerate the anaerobic digestion of sewage sludge," *Water Sci.Technol*. **36** (11), 121-128.
6. Shimizu, T, Kudo, K. and Nasu, Y. (1993) „Anaerobic waste activated sludge digestion – bioconversion and kinetic model. *Biotechnol. Bioengng*. **41**, 1082-1091.
7. Clasen, J. (2002) "Inactivation of plankton by ultrasound in drinking water treatment," In: *Ultrasound in Environmental Technology II*, ed. by Neis U., TUHH Reports on Sanitary Engineering**, 35**, Hamburg, ISBN 3-930400-47-2, 137-144.
8. Hua, I. and Hoffmann, M.R. (1997) „Optimization of ultrasonic irradiation as an advanced oxidation technology," *Environ.Sci.Technol*. **31**, 2237-2243.
9. Hua I. and Thomson J.E. (2000) "Inactivaton of Escherichia coli by sonication at discrete ultrasonic frequencies," *Wat. Res*. **34** (15), 3888-3893.
10. Cummins C.S. (1989) "Bacterial cell wall structure" In: *Practical handbook of microbiology*, ed. by O'Leary W.M., CRC Press, New York, 349-379.
11. Lezcano I., Pérez-Rey R., Baluja Ch. and Sánchez E. (1997) "Ozone Inaction of Pseudomonas aeruginosa, Escherichia coli, Shigella sonnei and Salmonella typhimurium in Water," *Ozone Sciences & Engineering.* **21,** 293-300.

oooOOOooo