Water disinfection by sonophotochemical method using persulfate

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Abstract. The efficiency of water disinfection was investigated using advanced oxidation processes (AOPs) based on ultraviolet (UV) LEDs and high-frequency ultrasound (US) in the presence and absence of persulfate (PS). The hybrid oxidation system {UV+US+PS} was found to be the most effective for inactivating *Enterococcus faecalis* in a row: {UV+US+PS}>{UV+PS}>{UV+US}>{UV}>{US+PS}>{US}. The effect of US in this hybrid system for *Escherichia coli* was not observed and the systems {UV+US+PS} and {UV+PS} were equally effective. Complete inactivation (100%) of *E. faecalis* and *E. coli* by sonophotochemical method using PS was achieved after 10- and 7-min treatment, respectively. We suppose that this method is promising for effective water disinfection.

1. Introduction

By present time, microbial pollution of aquatic ecosystems and deficiency of high-quality drinking water has become one of the global problems. Microbial pathogens come into natural waters as components of untreated or poorly treated wastewater. Therefore, it is necessary to use modern methods of water disinfection for reducing microbial contamination and providing drinking water quality. Traditional methods of disinfection (e.g., chlorination, ultraviolet (UV) irradiation) have disadvantages, such as formation of toxic byproducts and development of microbial resistance and reactivation. During last years, advanced oxidation processes (AOPs) for water treatment and disinfection have been more intensively studied. In view of environmental safety and energy-efficiency, the sonophotochemical method (UV+US), based on irradiation of aqueous medium by US and UV, can be considered as one of the most promising AOPs. Since UV light is absorbed by water containing suspended particles, an additional US treatment enhances the efficiency without further increasing UV dose [1]. Previously, the antimicrobial effects of low-frequency US (<100 kHz) in combination with UV radiation under sequential and simultaneous modes were studied [2-9]. For example, Blume and Neis (2004) demonstrated the efficiency of combined treatment of water by US and UV and found that *E. coli* concentration was decreased by 3.7 logs and power consumption was reduced by 57%. The synergistic effect of sequential treatment US→UV was observed for *E. coli* inactivation [8]. The elimination of total coliforms and *E. coli* from wastewater was studied [9]. In this work, the synergistic effect was also found using UV (254 nm) and low-frequency US (39 kHz). The decrease of *E. coli* number to 10 CFU per 100 mL was observed after 15 min and biofouling of lamps was also reduced. In the above studies, low-frequency US was used, whereas the antimicrobial effect of high-frequency US coupled with UV irradiation (> 100 kHz) was little studied.

It is known that highly reactive radicals, which are capable of inactivating of microbial cells, are generated upon photoactivation of PS. For example, *E. coli* inactivation was investigated by PS-activated visible light and this method was applicable for inactivating pathogenic bacteria *Staphylococcus aureus* and *Pseudomonas aeruginosa* [10]. In addition, post-treatment of water, contaminated with *E. coli* and *E. faecalis*, was possible by UV activation of peroxomonosulfate (PMS) and PS [11]. Furthermore, *E. coli*, *S. aureus*, *B. mycoides* and *Candida albicans* were inactivated by UV-A light-emitting diodes (LEDs) in the presence of PMS [12]. Photolytic activation of PMS by LEDs provided full inactivation of these microorganisms by using low doses PMS (0.1 mM) and neutral pH.

The hybrid {UV + US} treatments in the presence of oxidants or catalysts are promising to improve the rate and efficiency of inactivation. In particular, such processes, based on US (> 100 kHz) and UV irradiation in the presence of PS {UV+US+PS}, for water disinfection have not been studied before. The aim of this work was to study the efficiency of *E. faecalis* and *E. coli* inactivation in this sonophotochemical system.

2. Materials and methods

The bacterial strains of *Escherichia coli* K-12 and *Enterococcus faecalis* B 4053 were used as model test organisms; these are universal indicators of microbial contamination of water (State Research Institute of Genetics and Selection of Industrial Microorganisms of National Research Center, Moscow, Russia). Lyophilized cells were inoculated in nutrient broth: *E. coli* in GRM broth (State Research Center for Applied Microbiology and Biotechnology, Obolensk, Russia) and *E. faecalis* in trypticase soy broth (Merck, Germany). Cultivation was carried out aerobically in a shaker-incubator (Biosan ES-20, Latvia) overnight at 37°C and 180 rpm. Cells from one-day culture were precipitated by centrifugation at 4000 rpm for 5 min (Centurion Scientific, UK), washed twice and resuspended in phosphate buffered saline. The resulting cell suspension was added to deionized water (Simplicity® UVsystem, Millipore) for obtaining initial cell number (N_0) of ~ 10^5 colony forming units (CFU) per 1 mL.

The potassium persulfate was used as an oxidizing agent (Vekton, Russia) and its initial concentration was 312.5 μM [13].

The batch sonophotoreactor was comprised of UV LEDs, emitting at 365 nm (Yonton, model YT-100WUV370-0, China), and US generator with a frequency of 1.7 MHz (Scoole, model SC HR UL 04 (VO), China) (Figure 1). Water was irradiated under magnetic stirring (IKA® Color Squid white, Germany). The efficiency of inactivation was determined by comparing the mean number of CFU after control incubation (non-irradiated) and irradiated sample on agar medium for 24 h in triplicates. The obtained results are graphically represented as inactivation degree ($Lg(N/N_0)$) versus exposure time (min).

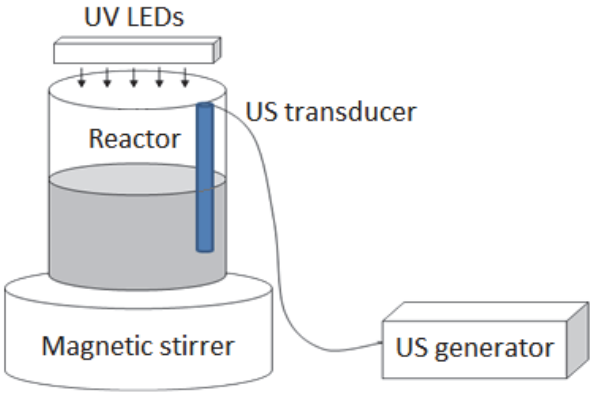

Figure 1. Schematic diagram of the used sonophotoreactor.

E. faecalis and *E. coli* were sequentially irradiated in the sonophotoreactor under the following conditions:

1. Irradiation with UV LEDs {UV};
2. Ultrasonication {US};
3. Simultaneous irradiation with LEDs and ultrasound {UV+US};
4. UV irradiation in the presence of persulfate {UV+PS};
5. Ultrasonication in the presence of persulfate {US+PS};
6. Simultaneous irradiation by LED and ultrasound in the presence of persulfate {UV+US+PS}.

3. Results and discussion

Figure 2 shows the survival curves obtained after {UV}, {US}, {UV+US} treatments, and in the same systems in the presence of PS.

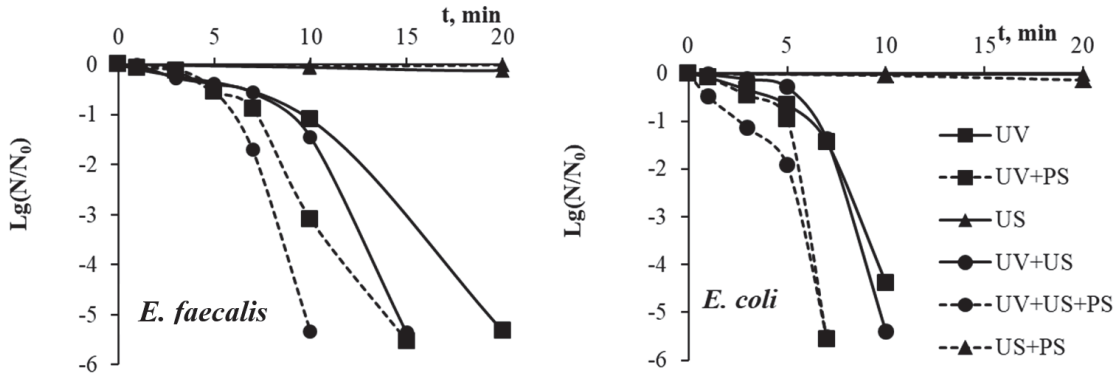

Figure 2. Inactivation *E. faecalis* and *E. coli* with an initial number of 10^5 CFU/mL in various oxidation systems using UV LEDs and US in the presence and absence of persulfate.

As shown in Figure 2, *E. faecalis* and *E. coli* by high-frequency US in the absence and in the presence of persulfate was not inactivated. The complete inactivation of *E. coli* and *E. faecalis* by direct UV irradiation was achieved after 10 and 20 min, respectively. It was found that the hybrid system {UV+US+PS} was the most effective for inactivating *E. faecalis* and shortened the exposure time to 10 min for achieving 100% inactivation (Figure 2). Presumably, this is due to higher yield of radicals ($SO_4^{\bullet-}$, OH•), generated by co-activation of PS by UV and US radiation.

Regarding *E. coli*, no visible difference between {UV+US+PS} and {UV+PS} systems was observed. However, less time (7 min) was required for complete inactivation, which indicates the higher sensitivity of *E. coli* cells to radicals attack. It is known that *E. coli* (a gram-negative bacterium) and *E. faecalis* (a gram-positive bacterium) are characterized by different cell wall structures [14]. Murein layer of gram-positive bacteria embedded mainly polysaccharides and proteins, which makes cell wall relatively thicker (20-80 nm). Its main component, peptidoglycan, is 40-90% by dry weight, and performs a forming and protective function. Gram-negative bacteria have thinner cell wall and a more complex structure. It consists of three layers: the outer - lipoprotein, the medium - lipopolysaccharide and the internal - peptidoglycan. The internal peptidoglycan layer (2-3 nm) is 5-10% by dry weight. The outer layer (8-10 nm) is composed of lipopolysaccharides, proteins, phospholipids and follows by a thin peptidoglycan layer (i.e., murein layer) [15].

A "plateau" on the *E. coli* inactivation curves was observed during the first 5 min treatment for {UV+US} system. The inactivation degree was about 0.5-0.6 log reduction. A similar effect for *E. faecalis* was observed for all systems within 5 min irradiation, resulting in 0.5 log reduction. However, no plateau effect was found for treating *E. coli* in {UV+US+PS} and {UV+PS} systems. This plateau can be explained by screening effect, when lower layers of cells are shaded by upper ones, requiring longer treatment times.

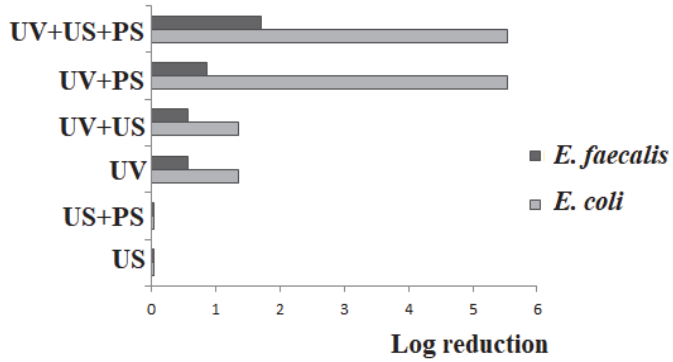

Figure 3. Comparative efficiency of inactivation of *E. coli* and *E. faecalis* in various systems using UV LEDs and high-frequency US without and in the presence of PS after 7 min of treatment.

Figure 3 shows that after 7 min treatment the hybrid system {UV+US+PS} was the most effective for *E. faecalis* inactivation, whereas the efficiency of *E. coli* inactivation by {UV+PS} treatment with and without ultrasound was comparable.

4. Conclusions

1. The oxidation system {UV+US+PS} was the most effective for *E. faecalis* inactivation.
2. *E. coli* was less resistant to UV and US exposure than *E. faecalis* due to different cell wall structure.
3. The obtained results demonstrated the applicability of hybrid sonophotochemical method using high-frequency US and UV LEDs for efficiency water disinfection.

Acknowledgments

This study was supported by the State Project of BINM SB RAS (No. 0339-2019-0005).

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