



Use of hydrodynamic cavitation in (waste)water treatment



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ABSTRACT

The use of acoustic cavitation for water and wastewater treatment (cleaning) is a well known procedure. Yet, the use of hydrodynamic cavitation as a sole technique or in combination with other techniques such as ultrasound has only recently been suggested and employed.

In the first part of this paper a general overview of techniques that employ hydrodynamic cavitation for cleaning of water and wastewater is presented.

In the second part of the paper the focus is on our own most recent work using hydrodynamic cavitation for removal of pharmaceuticals (clofibrilic acid, ibuprofen, ketoprofen, naproxen, diclofenac, carbamazepine), toxic cyanobacteria (*Microcystis aeruginosa*), green microalgae (*Chlorella vulgaris*), bacteria (*Legionella pneumophila*) and viruses (Rotavirus) from water and wastewater.

As will be shown, hydrodynamic cavitation, like acoustic, can manifest itself in many different forms each having its own distinctive properties and mechanisms. This was until now neglected, which eventually led to poor performance of the technique. We will show that a different type of hydrodynamic cavitation (different removal mechanism) is required for successful removal of different pollutants.

The path to use hydrodynamic cavitation as a routine water cleaning method is still long, but recent results have already shown great potential for optimisation, which could lead to a low energy tool for water and wastewater cleaning.

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1. Introduction

Availability of water is becoming an increasing concern in the globalised world, both in developed and in developing countries. A sustainable use of water sources could result in the search of additional water sources or even in recycling wastewater treatment plant effluents [1]. The goal of biological wastewater treatment is a stepwise oxidation of organic pollutants aiming to achieve complete mineralisation. Yet, numerous wastewater constituents are persistent to biodegradation or they are only subjected to minor structural changes instead of complete transformation into carbon dioxide and water. Alternatively, they may be eliminated by applying advanced abiotic treatment processes such as membrane filtration, UV degradation, ozonation, advanced oxidation processes, one of them being cavitation.

Cavitation, i.e. the appearance of vapour cavities inside an initially homogeneous liquid medium, occurs in very different situations. It can be defined as the breakdown of a liquid medium under very low pressures. This makes cavitation relevant to the field of continuum mechanics and it applies to cases in which the liquid is either static or in motion. When an oscillating pressure field is applied over the free surface of a static or nearly static liquid contained in a reservoir, cavitation bubbles may appear within the liquid bulk if the oscillation amplitude is large enough. This type of cavitation is known as acoustic cavitation. However, cavitation can also occur in a liquid, which is in motion. In liquid flows, this phase change is generally due to local high velocities, which induce low pressures. The liquid medium is then “broken” at one or several points of weakness (gas bubbles, impurities) and larger “voids” (bubble clouds) appear whose shape depends strongly on the structure of the flow. In the developed cavitation, which is the focus of the present study, the flow follows a distinctive pattern where cavitation structures of different shapes and sizes are shed

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from the attached cavity (Fig. 1a) – the flow is from the right to the left.

Developed cavitation occurs when the pressure difference between the outer flow and the inside of the attached cavity, forces the streamlines to curve towards the cavity and the surface beneath it. This causes the attached cavity to close and the formation of a stagnation point at which the flow is split into outer flow which reattaches to the wall and the re-entrant jet which travels upstream, carrying a small quantity of the liquid to the inside the cavity. As the re-entrant jet travels upstream it loses momentum, turns upwards and “cuts” the attached cavity, causing cavitation cloud separation (shedding). The cloud is then entrained downstream by the main flow and can violently collapse in a region of pressure recovery. During the separation, circulation around the structure can appear, causing it to reshape, break up etc. Meanwhile the attached cavity begins to grow and the process is periodically repeated.

As the system pressure is decreased or the flow velocity is increased a small cavity will extend and grow longer and longer. It becomes a supercavity as soon as it ceases to close on the cavitator wall but inside the liquid, downstream of the cavitator (Fig. 1b) – the flow is from the right to the left. Supercavitating flow shows only one quasi steady vapour filled large scale cavity, where larger disturbances in pressure and temperature are uncommon – it is not accompanied by noise, vibration and erosion, which would make the operation of a real facility somewhat easier.

Despite many obvious differences in appearance, on a small scale the principles which govern the hydrodynamic bubble and the acoustic bubble are basically the same. Once the cavitation bubble is generated, it may undergo a violent collapse during which an intense shock wave is emitted. Pressures up to a GPa range and high local temperatures, in the order of 10,000 K can be expected [2]. These conditions are uniquely suited for mechanical substrate surface or membrane cleaning, cell disruption or enhanced oxidation of chemical compounds.

With cavitation one utilises (i) extreme pressures and temperatures from cavitation collapses to disintegrate smaller organic molecules, which are otherwise harder to disintegrate using conventional biological methods and (ii) disintegration of larger particles to enlarge the specific surface and thus increase the rate of hydrolysis and biodegradation of organic pollutants.

Cavitation can be combined with conventional biological treatment using activated sludge. By decreasing the amount of persistent organic pollutants in wastewater treatment plant effluent, we will demonstrate the improved efficiency of treatment.

In the current industrial practice of wastewater treatment hydrodynamic cavitation is not used. Although laboratory experiments exist, the methods have not been routinely applied for practical use – some attempts include [3–6]. This is, in our opinion, mainly due to lack of communication between researchers – the environmentalists concentrate their efforts on ultrasonic cavitation, while the engineers do not realise the usefulness of cavitation and still treat it as a harmful phenomenon.

Hydrodynamic cavitation has the potential to become energy efficient technique that can reduce currently necessary use of expensive chemical reagents for enhanced treatment process, which on the other hand also pose additional concerns when deposited into environment. Cavitation as physical phenomena does not introduce any new chemicals to water and thus does not affect the environment after water is released into environment. Finally, as nowadays a lot of attention is put upon micropollutants such as endocrine disrupting compounds, it is expected that developed process of wastewater treatment with aid of cavitation will considerably reduce their presence in purified water. We expect that cavitation could also be used for disinfection of waste- and drinking water.

Also new applications of hydrodynamic cavitation are beginning to emerge in other fields i.e. decreasing the addition of sulphur in wine production, enhancing bio-gas production from waste activated sludge and homogenisation of pulp in paper production.

Part 1 (Section 2) of the present paper describes a review of methods, techniques and mechanisms, which utilise hydrodynamic cavitation in water and wastewater treatment with focus on pharmaceuticals, bacteria, microalgae and viruses removal.

In Part 2 (Section 3) of the present paper, results of research work performed by our project group during the last three years under the grant of the Slovenian Research Agency are shown. The goal of the research was to investigate the possibility and later energy efficiency of removal of different pollutants from water and wastewater with the final goal to develop an energy efficient industrial scale water and wastewater treatment facility, which would primarily utilise hydrodynamic cavitation for pollutant removal/disintegration.

2. Part 1: a review of methods and mechanisms

This section describes a review of recent applications where hydrodynamic cavitation is used for treatment of water and wastewater with the focus on pharmaceuticals, bacteria, microalgae and viruses removal.

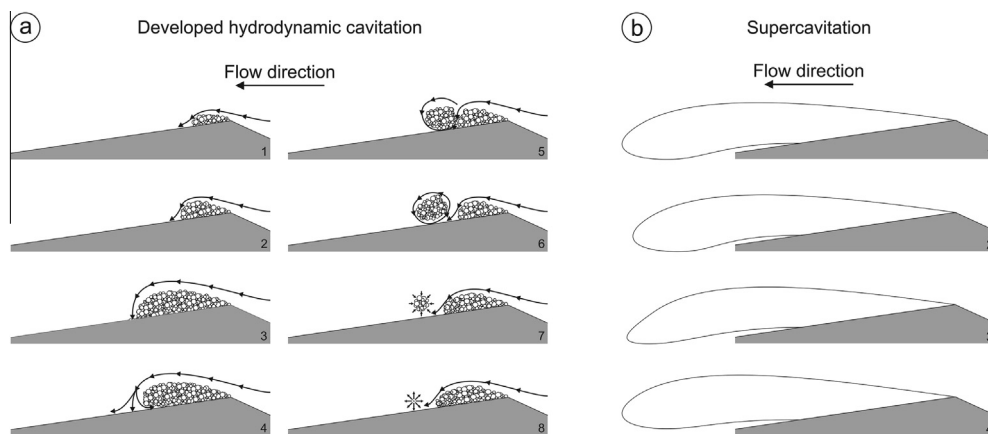


Fig. 1. Schematic representation of “developed hydrodynamic cavitation” (a) where highly dynamical vapour cloud shedding associated with high pressure pulsations is expected and “supercavitation” (b), which is characterised by a single quasi steady large cavitation pocket.

2.1. Pharmaceuticals

Pharmaceutical residues are nowadays acknowledged as an emerging environmental problem. They are produced for use in human and veterinary medicine and in animal husbandry [7]. In target organisms, most pharmaceuticals undergo metabolic transformation, which decreases their pharmacological activity and facilitates their excretion [8]. Due to metabolism, parent pharmaceuticals can be excreted as unchanged compounds, as a major metabolite or as a group of many different metabolites [8]. Since certain amounts of pharmaceuticals leave target organisms as unchanged compounds, they can enter into different environmental compartments and in parallel with increasing production and consumption, also the burden on the environment is increasing.

Pharmaceuticals can enter the environment through various routes (hospitals, households, unused medicines, animal excretion, etc.) [9], but generally wastewater treatment plant effluents are considered the most critical point source. Once in the environment their fate depends on their physico-chemical properties such as water solubility, volatility, octanol/water partition coefficient (K_{ow}), sorption constant (K_d) and dissociation constant (pK_a) [8].

Environmental concentrations of pharmaceuticals measured in various water compartments usually range from low ng L^{-1} to a few $\mu\text{g L}^{-1}$ [9] and although their concentrations vary, continuous input may result in detrimental effects on non-target organisms including humans. Researches also reveal the detrimental effects that these compounds can have on aquatic organisms [10]. These studies confirm the need to improve or upgrade conventional wastewater treatment.

As a precaution, and to ensure that throughout the EU a sufficient quantity of good quality water is available the Water Framework Directive was put in action in 2000. The first step was the establishment of a list of 33 priority substances in 2001, which were to be monitored and controlled in EU surface waters. Now the European Commission issued a Proposal for a Directive [11] amending the EU Water Framework Directive [12,13], where 15 additional priority substances are considered. For the first time three pharmaceuticals are included in the group of priority substances (diclofenac, 17β -estradiol and 17α -ethinylestradiol). When sufficient data are gathered, it is expected that other pharmaceuticals will follow.

2.1.1. Removal mechanisms

Certain pharmaceuticals like clofibrac acid, carbamazepine and diclofenac are resistant to elimination by conventional biological wastewater treatment and are therefore more likely to pollute the environment. To prevent entry and potential adverse effects of bio-recalcitrant compounds in the environment investigation of novel non-biological techniques as a precaution seems reasonable. Great effort has been put into investigating different chemical and photochemical advanced treatment processes such as photolysis [14], ozonation [15] and various advanced oxidation processes [16–18]. Most of these studies investigate the removal of only a few pharmaceuticals and not mixtures and also in matrices far less complex than wastewater. In addition, advanced treatment techniques like hydrodynamic cavitation are yet to be fully investigated for their potential to remove a mixture of pharmaceuticals from wastewater. Prior to our work, which is more thoroughly described in the second part of the paper only two studies existed where removal of pharmaceuticals was investigated using hydrodynamic cavitation [19,20]. These two studies indicated that hydrodynamic cavitation has the potential to efficiently remove bio-recalcitrant carbamazepine and diclofenac and show that studies reporting on removal of other pharmaceuticals are necessary. In addition, studies reporting on efficiency of hydrodynamic cavitation in more complex matrices are necessary in order to assess

the potential of hydrodynamic cavitation as a technique for removal of micropollutants from wastewater treatment plant effluents.

During hydrodynamic cavitation high local temperatures of 5000 K lead to the formation of different radicals (predominately $\text{OH}\cdot$ and $\text{H}\cdot$) after homolytic cleavage of water molecules [19]. Breakdown of organic compounds during hydrodynamic cavitation can occur at three locations (i) in the gas phase i.e., inside the bubble, where thermolytic decomposition of volatile compounds and $\cdot\text{OH}$ formation take place; (ii) at the gas–liquid interface, where degradation of non-volatile and hydrophobic compounds can occur, and (iii) in the liquid bulk phase, where degradation of non-volatile and hydrophilic compounds can take place [21]. Which of the mechanisms predominates depends on the properties of the compound and cavitation pattern and intensity [18]. Since only a small amount of radicals reach the liquid bulk phase, as they either react between themselves or with any oxidizable compound in the vicinity, removal of organic compounds depends on their physico-chemical properties [21]. To intensify hydrodynamic cavitation process and to improve removal of compounds found mostly in the liquid bulk phase, the addition of external oxidants e.g. hydrogen peroxide (H_2O_2) as a source of radicals is also an option [22,23].

Also the influence of temperature on the elimination of different organic compounds by cavitation has been investigated [19,23]. For different compounds (e.g. increasing temperature from 30 to 40 °C augmented the degradation of alachlor, while increasing the temperature to 60 °C resulted in decreased degradation; another study showed that increasing the temperature from 32 to 39 °C had no effect on methyl parathion degradation; a decrease in the degradation of dichlorvos was observed when increasing the operating temperature from 31 to 39 °C; a study on CBZ degradation using a combination of hydrodynamic and acoustic cavitation found that the temperature of 25 °C is optimal). These studies suggest that an optimal operating temperature needs to be empirically determined for a specific system in order to achieve the highest hydrodynamic cavitation efficiency. The effect of temperature on the degradation also depends on the investigated compound. Different removal efficiencies under identical operating parameters proved that the individual chemical properties of the compounds play an important role.

2.2. Bacteria

Although most bacteria are harmless or often beneficial, several are pathogenic. Pathogenic bacteria can cause infections such as tetanus, typhoid fever, diphtheria, syphilis, cholera, foodborne illness, leprosy and tuberculosis and are a major cause of human death. They can also contribute to other globally important diseases, such as pneumonia. Due to several recent outbreaks and the evident increase of reported cases of Legionnaires' disease (from 4.1 per million population in 1993 to 11.8 per million population in 2008 [24]) the efficient eradication of the bacteria *Legionella pneumophila* became a growing interest of many researchers.

L. pneumophila is wide spread in all natural fresh water sources in predominantly low concentrations. The bacteria has also frequently been observed in engineered water systems such as warm water distributing systems, cooling towers, humidifiers and fountains [25].

In low concentrations *L. pneumophila* does not represent a significant risk for the health of humans, however the multiplication of the bacteria in water systems poses a potentially fatal (between 15% and 20% of those infected [26]) human health risk wherever aerosolisation can occur [27].

Numerous measures can be used to create water systems in the built environment hostile to the multiplication of *L. pneumophila*

such as storage of cold water below 20 °C or storage of hot water above 60 °C [28]. When an outbreak occurs thermal shocks (raising water temperature at 70 °C for as much as 30 min) are most traditionally employed [29]. Other methods include shock chlorination disinfection, which involves injecting chlorine (50 ppm) into the water distribution system [30] and copper–silver ionisation [31].

The effects of cavitation, initiated by ultrasound, on bacteria are well known [32]. Acoustic cavitation is generally accepted as an effective method for sterilization, but one must also consider its limitations such as inability to treat larger volumes (operation in batch mode), poor scalability from lab to industrial scale and high operational costs [33].

On contrary, very few studies investigated hydrodynamic cavitation as a possible tool for bacteria eradication. Effect of hydrodynamic cavitation on disinfection of *Escherichia coli* was investigated in laboratory scale device by Mezule et al. [34]. The cavitation was generated using a rotor in the thin layer of water, which was circulated from and to a reservoir. Experiments showed that hydrodynamic cavitation was very effective in reducing bacterial ability to divide. Exposure of 3 min using energy input of 490 W/L stopped the division of 75% of *E. coli* cells.

Arrojo et al. [35] theoretically and experimentally studied hydrodynamic cavitation as an advanced oxidation process to understand the mechanisms involved in *E. coli* cell disruption. They obtained results contrary to those obtained by acoustic cavitation, where chemical processes caused by ·OH radicals seem to play a major role. Theoretical predictions and experimental observations have indicated that hydrodynamic cavitation disinfection, with comparatively slow pressure oscillations (low frequency), is mainly caused by mechanical disruption of bacteria. Thus, the disinfection rates are maximised by those configurations and operation parameters, which promote large bubbles, extended pressure oscillations and a larger number of cavitation events (i.e. conditions found in the Venturi tubes).

Loraine et al. [36] investigated cavitating jet technologies as means of disinfection of gram-negative *E. coli*, *Klebsiella pneumoniae*, *Pseudomonas syringae* and *Pseudomonas aeruginosa* and gram positive *Bacillus subtilis*. The hydrodynamic cavitation jets were found to be very effective in reducing the concentrations of all of these species and also the power efficiency of the process was shown to be superior to the acoustic cavitation. Balasundaram and Harrison [37] studied hydrodynamic cavitation for the partial disruption of *E. coli* and selective release of specific proteins relative to the total soluble protein. The effects of the cavitation number, the number of passes, and the specific growth rate of *E. coli* on the release of periplasmic and cytoplasmic proteins were investigated. At the optimum, 48% of the total soluble protein, 88% of acid phosphatase, and 67% of β-galactosidase were released by hydrodynamic cavitation in comparison with the maximum release attained using multiple passes through the French Press.

In general the described methods are only useful for mitigating the multiplication of planktonic bacteria, which are found in a free stream of the water system – they cannot be applied for the prevention of biofilm formation or destruction. To prevent the widespread of bacteria the process must therefore operate continuously, hence the energy efficiency of a method is of a decisive importance.

2.2.1. Removal mechanisms

The processes which accompany the phenomenon of the collapsing cavitation bubble such as high local temperatures of 5000 K which lead to the formation of different radicals (predominantly OH· and H·) and consequent oxidation, are likely not important for the eradication of bacteria due to the large sizes of the organisms [35]. On a bacterial size scale a more obvious removal mechanism are the shock waves, shear flow, super critical water

conditions, pressure and temperature spikes which accompany the aggressive bubble collapse. In addition, our recent study showed [38], that rapid decrease of pressure at the initiation of the bubble growth can play an important (sometimes decisive) role in damaging the bacteria.

2.3. Cyanobacteria and microalgae

Cyanobacteria and microalgae are among the most important organisms responsible for primary production, nutrient and energy circulation in water ecosystems [39]. Cyanobacteria are prokaryotic organisms without cell organelles, while microalgae are eukaryotic organisms with cellulosic cell wall and cell cytoplasm divided into compartments. Almost all planktonic cyanobacteria like *Microcystis*, *Aphanizomenon* and *Nodularia* have gas vacuoles, which regulate their position in water column and are sensitive to different physical environmental factors such as high pressure and shear forces [39,40].

In eutrophic standing waters concentration of cyanobacteria and microalgae biomass can sharply increase provoking algal bloom [41], which can cause several problems in e.g. water distribution, retention, cooling, and treatment systems, aquacultures, and natural standing waters [42–44]. Increased algal biomass is responsible for faster clogging of filters and pipes, high water turbidity, drop of dissolved oxygen concentration in standing waters at night, and local flora and fauna change [44]. Microalgae can also pass filters and enter the drinking water system, where they became food for bacteria and fungi, which can cause health problems [43,44]. Even more, some species of cyanobacteria such as e.g. *Microcystis aeruginosa*, *Planktothrix agardhii*, *Aphanizomenon flos-aquae*, *Anabaena flos-aquae* produce toxins, which may affect human and animal health [45,46]. Fish and shellfish can accumulate these toxins and represent additional health risk to consumers [45]. Cyanobacterial metabolites such as geosmin and 2-methylisoborneol can negatively change organoleptic properties of drinking water and can cause unacceptable taste and odour of drinking water [47,48]. In aquaculture, presence of cyanobacteria in fish tanks can cause repulsive odour or taste in fish and shellfish, which makes them unsuitable for sale causing local economic damage [48]. Among cyanobacterial blooms, *Microcystis* bloom is one of the most common [43,49], hence the research focus of this section is the removal of *M. aeruginosa*.

2.3.1. Removal mechanisms

Cyanobacteria and microalgae can be removed from water using chemical, biological, and/or physical treatment. The easiest and the cheapest method for their removal is chemical treatment. By addition of chemicals to water such as copper-based algaecides, herbicides, photosensitizers, and chemical flocculants algal blooms can be efficiently reduced [50]. However, chemical treatment has several disadvantages like (i) toxicity against non-target organisms; (ii) generation of secondary pollutants; (iii) introduction of heavy metals to the water and their accumulation in the environment [46,51]. Algaecides can cause lysis of algal cell wall and release of cell content to the environment [50], which can be especially problematic in the case of toxic cyanobacterial bloom presence because the toxins are released from the cells into a water body [50]. In such cases, additional treatment of water by activated carbon or powerful oxidants is needed to remove or destroy dissolved toxins [45,50].

Biological removal of cyanobacteria and microalgae, which comprises e.g. grazers, and phytoplanktivorous fish [44,52] is gaining importance, because it is more environmentally friendly method as chemical treatment. Biological removal of cyanobacteria and microalgae can occur naturally or by biomanipulation, with introduction of new algae eating species to the eutrophic water

bodies [53]. Biomanipulation is faster than natural establishment of algae eating communities and can selectively affect only phytoplanktonic organisms such as cyanobacteria and microalgae, compared to chemical treatment, which affects also other water organisms [52].

Physical methods for cyanobacterial removal mainly consist of acoustic cavitation and hydrodynamic cavitation [54,55]. Acoustic cavitation is in general successful method to counteract cyanobacteria growth [56], by which the gas vacuoles inside algae cells, that act as “nuclei” for acoustic cavitation, are disintegrated and they collapse during the “bubble crush” period, resulting in the settlement of cyanobacteria [56]. Effects of acoustic cavitation on cyanobacteria removal depends on frequency, intensity and sonication time [47]. Acoustic cavitation can also reduce concentrations of cyanobacterial toxins, which have been released in the water due to cell lysis [55]. However, in some cases like in aquaculture systems, a low intensity ultrasound device without cavitation, which is installed directly in the fish tank can be used to counteract algae growth [57,58]. According to [59] non cavitation ultrasound treatment cause efficient sedimentation of planktonic algae, however, microalgae species like *Dictiosphaerium pulchellum*, *Pediastrum boryanum* and *Scenedesmus obliquus* are “tolerant” to ultrasonic irradiation.

The effects of acoustic cavitation on cyanobacterial and microalgal removal have been studied extensively [56,60]. However, only a few studies investigated the effect of hydrodynamic cavitation on algal removal [51,54,61]. Hydrodynamic cavitation has similar effects on cyanobacteria as acoustic cavitation [51,54,61]. During hydrodynamic cavitation, formation of cavities is followed by immediate implosion of cavities in the liquid and rapid pressure changes [62]. Locally high temperatures, high pressure, and formation of hydroxyl radicals during hydrodynamic cavitation affect algal cells causing a collapse of gas vacuoles, a damage of photosynthetic apparatus, and membrane structures in the cells [51,54,61]. The collapse of gas vacuoles results in rapid sedimentation of the cells, while the damage in photosynthetic apparatus and cell ultrastructures inhibits algal growth, and leads to their death [54,61]. Effects of hydrodynamic cavitation on algae depend on hydraulic characteristics of cavitation tube, inlet pressure, cavitation number, algae concentrations, and treatment times [61]. According to Jančula et al. [51], hydrodynamic cavitation is more effective on removal of cyanobacteria than green microalgae *Chlorella*. The reasons are the different cell structure, presence of cellulose in cell wall, and absence of gas vacuoles in *Chlorella*, compared to cyanobacteria. This indicates good potential of hydrodynamic cavitation for selective cyanobacterial removal from water bodies.

2.4. Viruses

An important requirement for water to be compatible with human use is its biological safety level, i.e., its level of contamination with waterborne pathogens such as bacteria and viruses. Among viruses, the most significant waterborne ones are Rotavirus, Norovirus, Astrovirus, Sapovirus, Adenovirus, Hepatitis A and E and Enterovirus. Most of them are causing gastroenteritis [63]. The mentioned viruses are shed in high concentrations into water systems and can survive long time in such environment, from several weeks to several months, or even years in some cases. Even highly diluted enteric viruses still pose a threat to humans due to their extremely low infectivity dose. In some cases 10 ingested virus particles are enough to cause an infection [64]. For these reasons, and in order to prevent health safety issues, wastewater should be properly treated before being used in agriculture or any other human related use. However, despite wastewater treatment plants, infectious viruses are released daily into the environment through the discharge of treated water and biosolids

[65]. In particular scenarios, drinking water is also at risk of contamination with pathogenic viruses, i.e., natural disasters, water recirculation systems, bioterrorism, etc. The US environmental protection agency, in its ground water rule, has stipulated that methods ensuring a 4 log reduction of pathogenic viruses should be used for water treatment [66].

In order to remove the pathogenic viruses present in the water for human consumption, several disinfection methods are utilised or are being evaluated, such as, filtration based methods, chemical disinfection (chlorination, ozone treatment), physical disinfection (UV, high pressure treatment, high temperature treatments). Each of them has pros and cons, related with the cost and efficiency of the treatment. The addition of chlorine for example, which is the most widely used disinfection technique, results, upon reaction with organic compounds present in the water, in the formation of disinfection by-products. Such compounds may have undesirable side effects on a long time scale, such as, mutagenic and carcinogenic effects [67].

2.4.1. Removal mechanisms

The mechanisms for virus inactivation are different and depend on the used inactivation procedure. Different virus elements (genome, surface proteins) can be affected by different treatments (UV, temperature, high pressure, etc.), which ultimately disable essential viral functions (host recognition, genome injection, replication, etc.) [68]. Two of the methods that are known to cause virus inactivation are heat [69] and high pressure [70]. The use of hydrodynamic cavitation for water treatment, alone or in combination with other disinfection procedures, has been described several times during the last decade, but the studies mostly focus on indicator bacteria and there is no information on the impact of cavitation on viral integrity and infectivity. Taking into account that cavitation results in locally generated high pressure and temperature micro changes and formation of free radicals, its use for virus disinfection seems promising and worth to explore.

3. Part 2: recent developments by our project group

The work presented in this section was performed during the last three years by the project group of the J7-4265 project supported by the Slovenian Research Agency. The goal of the project was to investigate the possibility and later energy efficiency of removal of different pollutants such as pharmaceuticals, bacteria, microalgae and viruses from water and wastewater. The final goal is the development of an energy efficient industrial scale water and wastewater treatment facility, which would primarily utilise hydrodynamic cavitation for pollutant removal/disintegration.

3.1. Experimental set-ups and types of cavitation

Experiments were performed at the Faculty of Mechanical Engineering of the University of Ljubljana with three different hydrodynamic cavitation reactors described in the following subsections. Preparation of samples and evaluation of test results took place at various cooperating institutions (Faculty of Health Sciences of the University of Ljubljana, Faculty of Civil and Geodetic Engineering of the University of Ljubljana, National Institute of Biology and at Jožef Stefan Institute).

The optimisation of the cavitation conditions followed a simple approach that the most aggressive cavitation will work best. The aggressiveness of cavitating flow is mainly related to the kinetic energy, which is transferred to potential energy of the cavitation structure and further on to the energy (amplitude) of the pressure wave at the cavitation structure collapse. Following this reasoning

the highest possible flow velocity and pressure were used in experiments.

3.1.1. Pulsating hydrodynamic cavitation reactor

Operating the reactor in cycles allows a more accurate evaluation of the cavitation phenomena after the preset number of pulsations (cycles). The set-up (Fig. 2a) was used for detailed studies of how and to what extent the developed cavitation contributes to the removal/destruction of the pollutants. This is why a pump was not included in the test loop, but pressure was used to force the treated water from one reservoir to the other.

The pulsating hydrodynamic cavitation reactor shown in Fig. 2a, consists of a 3 way valve, two 2 L reservoirs, and a symmetrical Venturi pipe (Fig. 2c) with a constriction of 1 mm height and 5 mm width (30 mm long converging and diverging sections with 10° angles), connecting both reservoirs. It is operated in cycles. Water is introduced into the reservoir 1, while the reservoir 2 remains empty. By opening the valve, compressed air at high pressure (5 bar) flows into the reservoir 1 and forces water to flow through the Venturi constriction into the second reservoir, where constant pressure is maintained at 1 bar (cavitation number, calculated on the basis of the pressure differences lies at $\sigma = 1.24$). As the flow passes through the constriction, it accelerates, causing a drop in the static pressure, which results in cavitation. The 3-way valve is electrically controlled – when a signal that the reservoir 1 is empty is received, it closes and then opens the path for the compressed air to flow into the reservoir 2 and for water to flow in the opposite direction and consequently cavitation is achieved at the other side of the Venturi constriction.

Contrary to other reactors used in this study the pulsating cavitation reactor did not have a pump (the main heat source in the loop) installed. Hence the water temperature did not increase significantly and no temperature control was needed.

3.1.2. Continuous hydrodynamic cavitation reactor

The continuous hydrodynamic cavitation test rig, shown in Fig. 2b, uses a pump for circulating the fluid through the Venturi section (Fig. 2c) with the same dimensions as the one in the pulsating setup. It consists of a 2 L reservoir, heat exchanger, pump and the Venturi section. Cavitation extent can be adjusted by either varying the flow velocity (rotational frequency of the pump) or the system pressure (air pressure pipe). For the case of developed hydrodynamic cavitation the pressure upstream of Venturi was held at 5 bar, this way the same cavitation conditions as in the pul-

sating setup are achieved (cavitation number, calculated on the basis of the pressure differences again lies at $\sigma = 1.24$). To achieve a different type of cavitation, namely stabile supercavitation, both the upstream pressure and the flow velocity needed to be adjusted (cavitation number of $\sigma = 0.75$ was used for supercavitating flow regime – flow velocity 6.7 m/s (at the throat (cross-section of 1 mm height and 5 mm width) of the Venturi), upstream pressure 0.2 bar).

3.1.3. Shear induced hydrodynamic cavitation reactor

By observing the cavitation influence on pollutants (pharmaceuticals, bacteria, microalgae, viruses) in both, above described, Venturi configurations a pilot shear induced hydrodynamic cavitation reactor was designed. It consists of two facing rotors with special radial grooves (described in more detail later on and shown in Fig. 3 – right), where each one is spinning in the opposite direction (Fig. 3). The rotors (90 mm diameter) driven by electrical motors with power of 0.37 kW each have special geometry, which causes periodically repeating pressure drops. The rotating frequency of the rotors is approximately 2800 rpm which means taking into account their diameter, that local velocities can reach up to 26 m/s (comparable to the conditions in the Venturi set-ups).

The design of the rotors is shown in Fig. 3 (right). One of the rotors had 11 grooves and the other had 12 grooves, to avoid the resonance. The distance between the two facing rotors was 0.8 mm. The teeth on the first rotor were right angled and were separated by 7 mm deep and 10 mm wide grooves. The teeth of the second rotor were inclined at an angle of 8°. This way, when the teeth of the two rotors are aligned, the gap between them resembles the Venturi nozzle geometry. The Venturi shape geometry of the teeth causes a low pressure zone – if the pressure is low enough, the cavitation forms. Cavitation is present in three different regions (Fig. 3, right). In the gap between the rotor and the housing, where attached cavitation forms on the leading edge of the teeth. Bubbles shed from the attached cavitation can also be seen here. When the two grooves are aligned, cavitation forms in the gap between the rotors (2). Finally cavitation clouds also form in the Venturi gap between the aligned teeth (3). Very rapidly changing pressure field points to an aggressive cavitation process. The scale-up of the design was studied recently and it showed promising results (good scalability) for both smaller and larger rotor diameters.

The type of cavitation forming inside the hydrodynamic cavitation generator is the so-called shear cavitation where cavitation

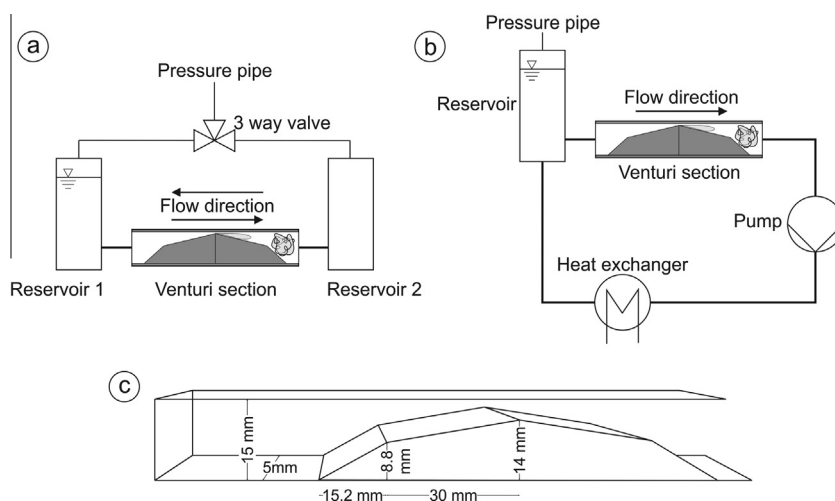


Fig. 2. Test set-ups for pulsating (a) and continuous (b) hydrodynamic cavitation reactors, with the small Venturi section (c) common to both set-ups. Adopted from [38,71].

investigate kinetics using a different setup in the future – we can however expect that the degradation of the compounds will follow pseudo first order kinetics as was already shown for carbamazepine [19] and diclofenac [20]. On the other hand changing the amount of added H₂O₂ showed that increasing the H₂O₂ dose positively influences the removal of pharmaceuticals, but only up to a point. For most pharmaceuticals, the optimal amount of added H₂O₂ was 0.34 g L⁻¹. However, when the amount of added H₂O₂ was increased to 6.8 g L⁻¹, the removal efficiency reduced for more than 15%. This is because, when H₂O₂ is in excess, it acts as a scavenger of ·OH [71,73]. Control experiments [72] showed significantly lower removal rates when only H₂O₂ was added (without cavitation). Similarly lower removals of pharmaceuticals were achieved without addition of H₂O₂ (cavitation only) which indicates that H₂O₂ enhances the removal of these compounds. Based on the physico-chemical properties of investigated pharmaceuticals [72] we can assume that the investigated pharmaceuticals are mostly found in the liquid bulk phase and are not in close proximity of the radicals that mostly appear at the gas-liquid interface. And since the concentration of radicals formed only by hydrodynamic cavitation bubbles is very low this could be the reason for low removal rates. However, with the addition of H₂O₂ the amount of radicals formed increases and can thus reach the bulk liquid phase and influence the removal of pharmaceuticals. Our results (Table 1) suggest that degradation of pharmaceuticals is driven by ·OH radicals generated from H₂O₂ under cavitation conditions. It is reported that H₂O₂ dissociates into ·OH under cavitation conditions [19,73] resulting in additional chemical oxidation, thus intensifying hydrodynamic cavitation efficiency, which was confirmed in our findings. Results also show that investigated pharmaceuticals are removed to a different extent (Table 1). Since destruction of organic compounds with hydrodynamic cavitation is dependent on their structure and chemical properties, the different chemical structure of the selected pharmaceuticals may be the reason for the different removal efficiencies observed.

Results for real wastewater effluents (Table 1) revealed that matrix composition reduces removal efficiency. Other organic and inorganic compounds present in the effluents, compete with the target compounds for ·OH radicals and thus affect the efficiency of the hydrodynamic cavitation process [74]. However, this effect can be compensated for by increasing H₂O₂ dose (3.4 g L⁻¹) and prolonging cavitation time (30 min). The results achieved under these conditions yielded comparable results to those achieved in deionised water and ranged from 37% to 79%. Even though the molar ratio between the amount of pharmaceuticals present in 1 L of real wastewater sample and H₂O₂ dose used to treat the sample (calculated from Table 1 last column on the right; pharmaceuticals: H₂O₂ = 3 × 10⁻⁷; 1) seems very high, one must be aware that this is the result of matrix complexity (besides pharmaceuticals the effluent sample also included a high amount of other organic and inorganic compounds, which, as already mentioned, competed with the target compounds (pharmaceuticals) for ·OH radicals).

3.3. Bacteria

In the present study the removal efficiency of *L. pneumophila* by three distinctive types of cavitation (i) the most commonly used acoustic cavitation, (ii) the developed hydrodynamic cavitation and (iii) the supercavitation were evaluated. The continuous hydrodynamic cavitation set-up (see Section 3.1.2, Fig. 2b) was used for (ii) and (iii). In addition, it was also tested whether the bacteria could be destroyed only by the non rapid exposure to low pressure – by evacuation of air from the flask by a vacuum pump – evaporation (low pressure boiling) occurs in about 10 s. For a more detailed description of the methods, see Šarc et al. [38].

Each of the three above mentioned cavitation types influence the bacteria in a specific way. In the acoustic cavitation the voids appear due to the tension in the liquid produced by an ultrasonic transducer. The bubble collapses usually follow the driving frequency of the transducer; hence one can anticipate a very intense cavitation dynamics. Most commonly, the high pressures and the temperatures, which occur at the bubble collapse, are mentioned as the driving mechanisms for water purification [75]. One of the drawbacks of the acoustic cavitation is that the bubbles appear only at antinodes of the standing waves in the vessel and some kind of mixing is therefore needed to sonicate the whole volume.

The concentration of energy at hydrodynamic cavitation cloud collapse results in high pressures and high local temperatures, which potentially damage the bacteria. Although more intense, the cavitation cloud collapses are less frequent than in the case of acoustic cavitation as they occur in the range of up to only a few hundred Hz. Consequently exposing the whole amount of fluid to cavitation is again an issue. Finally, the developed cavitation is known as the most aggressive one in terms of vibration, noise and erosion, which makes an actual bacteria removal facility difficult and expensive to run.

The bacteria could also be harmed in a regime of supercavitation if they are rapidly exposed to a very low pressure (i.e. vapour pressure, approximately 2000 Pa for cold water). The present test-section was designed in a way that the supercavitating bubble entrained the whole flow cross-section (that all the fluid underwent vapourization and condensation).

Measurements according to ISO 11731:1998 [76] standard were employed for enumeration of *L. pneumophila* organisms. Since the method is suitable for waters with prospected low numbers of *L. pneumophila* the sample was first diluted up to 10⁻⁵. 1 mL of diluted sample was spread onto buffered charcoal yeast extract agar and left to incubate at 36 ± 1 °C. Buffered charcoal yeast extract agar is a selective growth medium used to culture or grow only certain bacteria, particularly the gram-negative species of *L. pneumophila*. To determine the viable bacterial number the bacterial cells grown on the agar after the incubation were counted. Count of viable cells from 1 mL sample gives us estimation of concentration of colony forming units per volume CFU L⁻¹ (or CFU mL⁻¹).

Since we had no way of controlling the viable bacterial number in the prepared sample, it varied from 15,000 to 180,000 colony forming units mL⁻¹. In addition, no dependency between the removal rate and the initial (untreated sample) number of viable bacteria was found, hence to be able to compare the experiments, the measured values after the treatment were normalised by the measured viable bacterial number in the sample that was not exposed to treatment. For further analysis, the removal rate (RR) for each test was calculated as:

$$RR = \frac{([\text{CFU/mL}]_{\text{pre}} - [\text{CFU/mL}]_{\text{post}})}{[\text{CFU/mL}]_{\text{pre}}} \cdot 100 \quad (1)$$

where “pre” denotes CFU count prior to cavitation treatment and “post” the CFU count after the exposure to cavitation. The bacteria removal rate (RR) is then given in terms of % min⁻¹.

Results imply that a considerable difference in efficiency of bacteria eradication exists between the different types of cavitation (in addition to developed and supercavitating regime also acoustic cavitation and low pressure boiling were tested for their removal rates). Fig. 4 shows a diagram of removal rates achieved in a number of test cases for each cavitation type and the low pressure boiling.

Analysis of variance (ANOVA) showed statistically significant differences in bacterial removal rate among 4 sets of applied measurements at a confidence level higher than 99.9%. This undoubt-

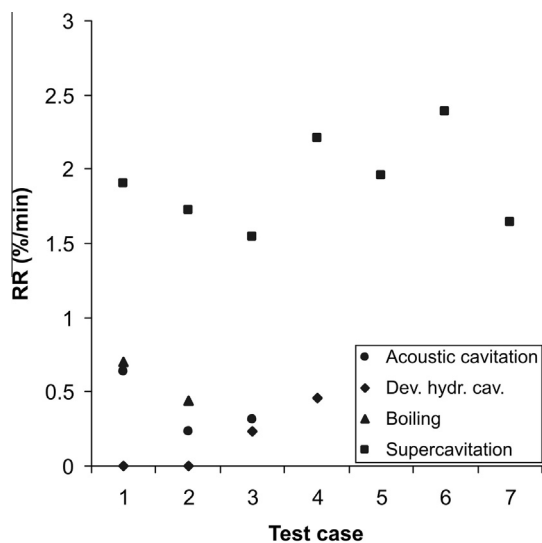


Fig. 4. Removal rates (RR) of *Legionella pneumophila* for each cavitation type and low pressure boiling.

edly confirms our reasoning that the bacterial removal rate in sample exposed to supercavitation is much higher than in other types of cavitation or boiling.

The physics behind the eradication of bacteria by supercavitation seems similar to cell disruption by nitrogen decompression [77]. The pressure drop at the transition from the liquid to vapour phase in the present case is almost instantaneous, as is the pressure rise at the cavity closure, which occurs downstream roughly 1 ms later. More importantly in the case of supercavitation, the treated volume is greater and the method can operate continuously rather than in batch mode, hence it is applicable for “real” water treatment procedures.

The fact that the low pressure boiling performed better than either acoustic cavitation or developed hydrodynamic cavitation shows, that it is the pressure decrease that damages the bacteria. In both acoustic cavitation or developed hydrodynamic cavitation we are predominantly dealing with pressure shocks which are emitted at single bubble and bubble cloud collapses. When this pressure decreases, it is at the same time also very rapid – in the order of a millisecond (like in the case of supercavitation), the efficiency of eradication increases to a level that could possibly be sufficiently high for an industrial application. This can be done within a reasonable amount of time in an average internal water supply system where recirculation of the hot water supply is installed.

The implementation of acoustic cavitation into a practical system was already pursued by many companies, but it has, as already mentioned, a deficiency that it is energy consuming and hard to develop in a continuous flow (not batch) mode. The hydrodynamic type of cavitation is easier to scale up and, by definition, operation is continuous. One possibility of implementation is the shear induced cavitation reactor described in Section 3.1.3. To generate supercavitation the geometry of the rotors would need to be modified and work is currently under way to achieve this and to test it in a model of hospital water pipe system. Based on our previous experience from other fields (pharmaceuticals), we are optimistic that the bacteria removal rate will increase when we make the transition from the Venturi design to the shear induced supercavitation reactor.

3.4. Cyanobacteria and microalgae

In the present study, the effect of hydrodynamic cavitation on toxic strain of cyanobacteria *M. aeruginosa* and a strain of green

microalgae *Chlorella vulgaris* was tested. *M. aeruginosa* culture (PCC 7806) was obtained from National Institute of Biology, Slovenia; while *C. vulgaris* culture was obtained from AlgEn, algal technology centre, Slovenia. Before the experiment, *M. aeruginosa* and *C. vulgaris* were cultivated in Erlenmeyer flasks at room temperature 23 °C in blue-green-11 medium and Bold’s Basal medium [78], respectively. Both cultures were illuminated with Flora lights (35 W/77, Osram, Germany) in 16/8 h light/dark intervals. To provide appropriate amount of sample for the conduction of the experiments, the cultures were diluted before the experiments with fresh nutrient medium blue-green-11 for *M. aeruginosa* and Bold’s Basal medium for *C. vulgaris* in ratio 1:1. After dilution initial cell concentration of test samples *M. aeruginosa* was 1.0×10^6 cells mL⁻¹; and 3×10^6 cells mL⁻¹ for *C. vulgaris*. The experiments were performed in at least four replicates. 300 mL of diluted *M. aeruginosa* and *C. vulgaris* samples were used as a control.

The experiments were performed with the use of continuous hydrodynamic cavitation reactor (see Section 3.1.2, Fig. 2b). The samples were exposed to hydrodynamic cavitation for 25 cycles (25 passes through the Venturi constriction). Before and after the hydrodynamic cavitation treatment temperature, pH, electric conductivity, and optical density at 684 nm were measured and cell concentration was determined according to Rodriguez et al. [79]. The UV–vis absorbance spectrum (400–700 nm) and observation of the samples with light microscope was performed to see potential structural changes of pigments and cell damage after the treatment. Optical density at 684 nm was measured due to the strong peak of chlorophyll-*a* in cyanobacterial and microalgal cells. Temperature, pH and electric conductivity were measured with EC/TDS/T tester HI98311 (HANA instruments, USA), optical density with Nanocolor UV–vis spectrophotometer (Macherey-Nagel, Germany). Algal cells were observed with light microscope Eclipse 80i (Nikon, Japan). For growth inhibition rate determination of 150 mL of test samples (after hydrodynamic cavitation exposure) and 150 mL of control samples were diluted each with 100 mL of fresh nutrient medium blue-green-11 (for *M. aeruginosa*) and Bold’s Basal medium (for *C. vulgaris*). The cultivation followed the same procedure as before the experiment (see previous paragraph). Absorbance of the samples was monitored for 6 days after exposure and cell concentration was determined based on calibration curve. Growth inhibition rate (GIR) was calculated as [63]:

$$\text{GIR} = \left(\frac{[\text{CellNum}/\text{mL}]_{\text{pre}} - [\text{CellNum}/\text{mL}]_{\text{post}}}{[\text{CellNum}/\text{mL}]_{\text{pre}}} \right) \cdot 100, \quad (2)$$

where “pre” denotes number of cells (*CellNum*) prior to cavitation treatment and “post” the number of cells after the exposure to cavitation. The growth inhibition rate is given in terms of %. After exposure of samples to hydrodynamic cavitation, no change in cell concentration of *M. aeruginosa* was noticed, while slight increase of absorbance at 684 nm was noticed in *C. vulgaris*. This is probably due to the disintegration of *C. vulgaris* aggregates into single cells during hydrodynamic cavitation treatment. Increase of absorbance in *C. vulgaris* samples after hydrodynamic cavitation is in accordance with results reported by Joyce et al. [80] and Jančula et al. [51].

Sedimentation of *M. aeruginosa* cells was observed 1 day after hydrodynamic cavitation treatment, while no cell sedimentation was present in control samples. Sedimentation of *M. aeruginosa* cells indicated damaged gas vacuoles in the cells resulting in inability to regulate cell position in the water column (inability of flotation). Xu et al. [61], Jančula et al. [51] and Li et al. [54] detected damage of gas vacuoles and change of *M. aeruginosa* cell structures after the hydrodynamic cavitation treatment. Sedimentation and damaged gas vacuoles were observed also in experiments with acoustic cavitation of cyanobacterial samples

[56,60,81]. No sedimentation was observed in treated *C. vulgaris* samples most probably due to the absence of the gas vacuoles in the cells of *C. vulgaris*.

Growth inhibition test for *M. aeruginosa* showed 50% rate in 2 days after hydrodynamic cavitation and 90% rate in 4 days after hydrodynamic cavitation (Fig. 5). The growth inhibition was increasing with time since hydrodynamic cavitation did not immediately cause the death of *M. aeruginosa* cells but it very likely caused the collapse of gas vacuoles in cells, which prevented the cells from floating and eventually resulted in the death of algae. According to these results, hydrodynamic cavitation is a technology that could effectively inhibit the growth of *M. aeruginosa*.

No growth inhibition was detected for *C. vulgaris*. According to our results the rate of *M. aeruginosa* was faster compared to the results obtained by Xu et al. [61], where *M. aeruginosa* was exposed to similar hydrodynamic cavitation setup for 8 cycles with 50% growth inhibition rate in 3 days and 64% in 6 days after hydrodynamic cavitation, indicating that for faster cyanobacteria removal more hydrodynamic cavitation cycles should be employed.

No changes in *M. aeruginosa* and *C. vulgaris* cells were observed after hydrodynamic cavitation treatment (light microscope), likewise a comparison of absorbance spectrum of the treated and the control samples for both species revealed no differences. This indicates that in both species, cell membrane maintained their function and integrity and that cell content was not released from the cells after exposure to hydrodynamic cavitation. This is positive in the case of toxic cyanobacterial blooms, where hydrodynamic cavitation is inhibiting their growth while toxic compounds are not released into the environment due to the cell lysis absence [51].

Our results showed that hydrodynamic cavitation treatment was not efficient for *C. vulgaris* removal. This confirms findings of Jančula et al. [51] that hydrodynamic cavitation can be a selective method for cyanobacterial bloom removal in water bodies. Such result can be seen as an advantage in lakes, ponds or reservoirs where it is desirable that green microalgae are not affected by the cavitation and may still act as the natural nutrient competitors of cyanobacteria [51]. In various water systems, where an efficient removal of all algae is necessary, the hydrodynamic cavitation can be combined with other treatment methods like UV irradiation, non-cavitation ultrasound or even chemicals [55,71].

Based on our results hydrodynamic cavitation alone or possibly in combination with other techniques has great potential for removal of cyanobacteria and microalgae from water (a techno-economic feasibility study would be needed to confirm this). Our results revealed the efficiency of hydrodynamic cavitation in

removal of cyanobacteria, however more research and improvements are needed, with the focus on the design of cavitation chamber to obtain higher pressure differences for the efficient microalgae removal.

3.5. Viruses

The effects of hydrodynamic cavitation on waterborne viruses have not yet been reported. In our ongoing study using a pulsating hydrodynamic cavitation reactor (Section 3.1.1) hydrodynamic cavitation is being evaluated as a step in water treatment for reduction of the presence of waterborne enteric viruses.

A Rotavirus clarified suspension was derived from routine Rotavirus positive clinical stool samples collected at the Institute for Microbiology and Immunology, University of Ljubljana, Slovenia. One litre of tap water was spiked with 0.5 mL of Rotavirus suspension. Spiked sample was treated using Venturi cavitation chamber with a pulsating system (Section 3.1.1, 400 passes through Venturi chamber). For detection and relative quantification of RoV, Rotavirus RNA was detected using RoV reverse transcription quantitative polymerase chain reaction (RT-qPCR) assay developed by Gutierrez-Aguirre et al. [82].

Preliminary results of measured difference between spiked sample before and after cavitation, suggest a promising performance of hydrodynamic cavitation with a 75% reduction of the detected RoV genomic RNA achieved with the cavitation treatment of the sample.

Additional experiments are in progress in order to draw conclusions for the efficiency of waterborne virus removal by hydrodynamic cavitation.

4. Conclusions

In the first part of the paper, we showed that the idea of using cavitation in water and wastewater treatment is not new. There is however a large gap between the existing applications that utilise acoustic cavitation and hydrodynamic cavitation. The widespread application of acoustic cavitation and relatively new idea of utilising hydrodynamic cavitation is, in our opinion, mainly a result of traditionally poor communication between the scientific fields in the past.

The second part of the paper shows a part of the work that was performed during the last few years by our project group. For the achievements, such as efficient eradication of *L. pneumophila* and high removal of various pharmaceuticals and cyanobacteria, the

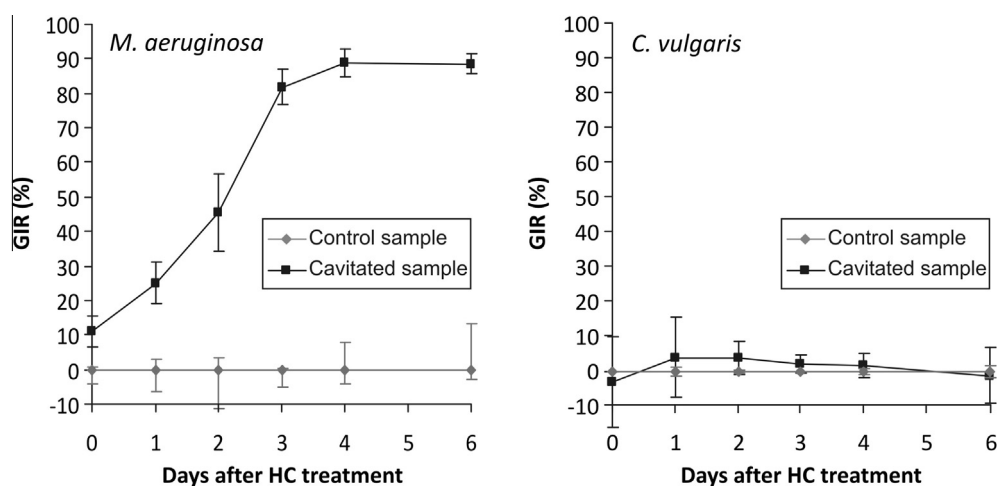


Fig. 5. Growth inhibition rate (GIR) of *Chlorella vulgaris* (right) and *Microcystis aeruginosa* (left), after hydrodynamic cavitation (HC) treatment.

essential piece of the puzzle was working in a very heterogeneous group of researchers.

Our recent investigations [38,71,72,83,84] point to the possibility that hydrodynamic cavitation is suitable for real applications as it is easily scaled, robust, it can operate in continuous mode and has in many cases higher removal efficiency than acoustic cavitation. However, a techno-economic feasibility study on a full scale reactor is still needed for employment of the technology into water treatment practice.

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