

MOGUĆNOST PRIMENE ULTRAZVUČNE KAVITACIJE U PROCESU PRERADE INDUSTRIJSKIH OTPADNIH VODA

POSSIBILITY OF USING ULTRASONIC CAVITATION IN THE PROCESS OF INDUSTRIAL WASTEWATER TREATMENT

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Sažetak

Razvojem industrije i rastom populacije tokom poslednjih godina javljaju se problemi u vezi sa otpadnim vodama iz proizvodnje. Sve otpadne vode iz postrojenja, bez obzira kako su nastale, sadrže štetne materije koje mogu da imaju i određen stepen toksičnih primesa i moraju se prečistiti ako njihov kvalitet ne odgovara zakonskim okvirima. Cilj tretmana otpadnih voda je uklanjanje štetnih materija čime se postiže smanjenje stepena zagađenosti. Većina zagađujućih materija su organskog porekla i njihovo uklanjanje se može realizovati primenom naprednih tehnika ultrazvuka i ultrazvučne kavitacije, a sve u cilju postizanja kvaliteta vode u skladu sa zakonskom regulativom. Princip delovanja ultrazvuka u obradi otpadnih voda temelji se na pretvaranju električne energije u vibracije određene amplitude i frekvencije, koje izazivaju razgradnju i uništavanje mikroorganizama i bakterija. Unapređeni procesi predstavljaju najefikasnije metode za uklanjanje i degradaciju teško rastvornih zagađujućih materija iz otpadnih voda. Proces prečišćavanja je optimizovan u odnosu na tehnološki postupak, koncentraciju, vreme tretmana i količinu primenjenog oksidacionog sredstva.

Ključne reči: *otpadna voda; ultrazvuk; kavitacija; kavitacioni mehur; zaštita životne sredine*

Abstract

With the development of industry and population growth in recent years, there are problems related to wastewater from production. All wastewater from the plant, regardless of how it was generated, contains harmful substances that may have a certain degree of toxic impurities and must be treated if their quality does not meet the legal framework. The goal of wastewater treatment is to remove harmful substances, which reduces the degree of pollution. Most of the pollutants are of organic origin and their removal can be realized by applying advanced techniques with ultrasound and ultrasonic cavitation to achieve water quality that complies with legal regulations. The principle of operation of ultrasound in wastewater treatment is based on the conversion of electricity into vibrations of a certain amplitude and frequency, which cause the decomposition and destroying of microorganisms and bacteria. Improved processes are the most efficient methods for removing and degrading sparingly soluble pollutants from wastewater. The purification process is optimized in regard to the technological procedure, concentration, treatment time and the amount of oxidizing agent applied.

Key words: *wastewater; ultrasound; cavitation; cavitation bubble; environmental protection*

1 INTRODUCTION

Technological wastewater is all wastewater discharged from the premises used to perform any commercial or industrial activity other than sanitary wastewater. Wastewater from the production process has proven to be very toxic to watercourses and the environment. Before release into the environment, it is necessary to process and remove toxic and biodegradable substances. Industrial wastewater originates from the production process or plant itself, washing raw materials, equipment

and cleaning warehouses, etc. These waters can pose a serious environmental problem if uncontrollably discharged into the environment. There are various physical, chemical and biological wastewater treatment processes, with their advantages and disadvantages.

The research is aimed at the development of new technologies in wastewater treatment and the reduction of harmfulness and toxicity by advanced electrochemical and oxidative processes, which have shown high performance in the removal of degradable substances. The application of ultrasound in wastewater treatment causes the decomposition and destroying of microorganisms and bacteria. Awareness of wastewater treatment and disposal is currently focused on research and application of ultrasound in environmental protection and the use of cavitation procedure to destroy chemical and biological substances in wastewater [1,2].

2 CAVITATION

Cavitation (lat. cavus - empty, hollow), as a phenomenon, was researched only at the end of the XIX century, and the first scientist who began to deal with cavitation was John William Strutt – 3rd Baron Rayleigh, an English physicist. Cavitation is a phenomenon that involves the formation and implosion of bubbles in a flowing fluid. Bubbles implosion leads to many undesirable phenomena such as increased pressure and temperature in the area of implosion, chemical corrosion and electrochemical processes, and surface damage to the material in contact with the fluid (cavitation damage).

Tests are most often performed in laboratory conditions, and field tests are generally not techno-economically justified, and due to the high cost, they are performed very rarely.

The following are used as laboratory methods for testing cavitation:

- vibrating;
- Venturi pipe method;
- rotating disk;
- liquid jet impact test method.

These are quite simple, fast (4 - 10 h) and cheap methods, often applied in engineering practice. In order to start studying the phenomenon of cavitation, it is necessary to apply a multidisciplinary approach, that is, to observe the process from the aspects of fluid mechanics, material engineering, thermodynamics, and technological operations [3].

2.1 *The principle of ultrasonic cavitation*

Ultrasound is a sound wave with a frequency higher than the upper limit of human hearing (20 kHz). There are three frequency ranges of ultrasound: relatively low frequency range (20 - 100 kHz), medium frequency range, (100 - 2000 kHz) and high frequency range (2 - 10 MHz). The effect of ultrasound treatment is basically a consequence of ultrasonic cavitation using sound ranges in the lower frequency range. The application is mostly in the range of 20 - 40 kHz, but there is a growing interest in the rise of higher frequencies, but not above the frequencies of MHz where cavitation is difficult to achieve without the use of high power.

The principle of action of ultrasound in wastewater treatment is based on the conversion of electricity into vibrations of a certain amplitude and frequency. When this energy is transferred to the sample, there are significant changes in its physical and chemical properties. After the absorption of ultrasonic energy, microscopic bubbles are formed in the sample, which grow during further ultrasonic irradiation and eventually the bubble bursts (explosion). Due to the implosion of cavitation bubbles, high temperatures and pressures develop in the immediate vicinity of the bubble (approximately 5000 K and 1000 bar, with a rapid temperature change of about 1010 K/s during the supply and removal of heat), thus causing an increase in chemical reactivity and free radicals.

The formation of bubbles, growth and explosion during the action of ultrasound on the sample is called ultrasonic cavitation. Ultrasonic waves are mechanical vibrations that propagate through a medium having elastic properties such as solid, liquid, or gaseous state.

The effect of ultrasound on the sample alternates between compression and expansion. In the expansion part of the cycle, ultrasound acts on the molecules in the sample and moves away from each other, and if the stress exceeds the strength of the sample, a cavity will form. Cavities are mostly formed in weak places in the sample, the so-called cavitation cores - gas-filled cracks.

The expansion part of the ultrasonic field causes a sudden growth of the formed cavities, during which they are formed by steam and fill the cavities with gas (bubbles). Once created, small gas bubbles will absorb sound energy and grow with further ultrasonic irradiation.

Bubble growth depends on the intensity of the ultrasound. Under the action of high-intensity ultrasound, a small bubble can grow very quickly, and if the expansion of the cavity takes place fast enough during the expansion, the bubble will not have enough time to re-compress.

At lower intensities, cavity growth also occurs, but with a slower process called rectification diffusion. Under such circumstances, the cavity will vary in size during expansion and compression. The growth of the bubble is slightly higher during each individual expansion than its compression during compression. The cavity after alternating cycles of expansion and compression can finally reach a critical size at which it can absorb ultrasonic energy. The critical size depends on the sample and the frequency of the ultrasound.

The bubble, if in the phase with the ultrasonic field, can grow and at one point become too big and can no longer absorb energy ultrasonic. In a field without energy intake, the bubble can no longer withstand and it bursts.

The development of enormously high local temperatures and pressures, as well as the achievement of extremely high rates of heat supply and removal during cavitation collapse, contributes to the use of ultrasound and enables unusual mechanisms for the formation of high-energy chemical reactions. For this purpose, the use of ultrasound can be used to break down the chemical bonds of pollutants in industrial wastewater [3-8].

2.2 Cavitation procedure in wastewater treatment

Regardless of the application of ultrasound, there are two basic ingredients, namely liquid media and a source of high energy vibration. Ultrasound has more and more potential for wastewater treatment. Chemical oxidation is used, which causes cavitation, the formation, growth and subsequent breakage of bubbles or cavities, formed in an extremely short time interval (microseconds), and the release of a large amount of energy.

Ultrasonic radiation destroys contaminants in water due to high concentrations of oxidizing species such as hydroxyl radicals and hydrogen peroxide. The growth and implosion of bubbles are influenced by the physical properties of the gas and the liquid, the size of the nuclei present in the liquid and the ultrasonic frequency and intensity. After the bubble breaks, the temperature can reach several thousand degrees. Under these conditions, organic compounds are degraded directly by pyrolytic cleavage. On the other hand, hydroxyl radicals formed by pyrolysis also participate in the decomposition of organic matter. The most common method used to create cavitation in the laboratory is ultrasonic cavitation.

The application of ultrasound in wastewater treatment causes the decomposition and destroying of microorganisms and bacteria. The oxidative action of free radicals formed in cavitation further accelerates ultrasonic processing. It is considered that microorganisms that survive the sample processing by ultrasound, unlike microorganisms after treatment with UV radiation, are not susceptible to genetic mutations. Ultrasonic irradiation of a sample causes high-energy chemical reactions that are often accompanied by light emission. Compression of the bubbles formed during ultrasonic cavitation is faster than heat transfer, and short-term "hot spots" in the sample are formed locally.

The primary process within the cavitation bubble is the formation of hydroxyl and hydrogen radicals in the hydrolysis of water molecules at high temperatures and pressures. OH- radicals can migrate to the boundary area and react or leave the boundary area and end up undisturbed. Equally, some pollutants can penetrate the cavitation cavity and undergo pyrolytic decomposition at extremely high temperatures and pressures. Under extreme conditions, the density, viscosity and ionic strength of water change. Under these unique conditions, water is a supercritical fluid and oxidation by OH-

Generated radicals can react with each other to form new molecules or new radicals, or they can diffuse into an undisturbed sample and serve as an oxidizing agent.

Approximately only 10 % of the hydrogen and hydroxyl radicals formed by the pyrolysis of water vapor reach the gaseous phase where they can enter the redox reaction. As the generation of radicals takes place at extremely high temperatures, it can be assumed that at some maximum temperature during the collapse of the cavitation bubble, most radicals will be formed. The final concentration of the formed radicals during the collapse of the bubbles depends on the physical characteristics. It is expected that the application of high-power ultrasound in aqueous samples saturated with argon will generate the most OH- radicals, because at higher cavitation temperatures more intensive homolysis of water molecules takes place.

Water dissociation occurs due to the thermal reaction caused by the last step of cavitation bubble collapse. Radical formation occurs after ultrasonic action and oxidative processes occur. The release of energy in the bubble is enough to dissociate water into H and OH radicals, but it is not enough to dissociate molecular oxygen.

The formation of hydrogen peroxide can only occur from the reaction between hydrogen atoms and molecular oxygen by cavitation:



The reaction takes place in two steps. In the first step, radicals are produced inside the bubble, while in the second step they migrate to the surface of the bubble where hydrogen peroxide reacts with phenol compounds. Lower frequencies are very successful in breaking down molecules inside bubbles. As the frequency increases, the destruction of the bubbles takes place faster and more radicals will come out of the bubbles [3-8].

2.3 Hydrodynamic cavitation reactors

Cavitation is considered as an effective and promising technology that has attracted the interest of many researchers and has been widely employed in many fields including disinfection, cell disruption, sludge treatment, bio-diesel synthesis, nano-emulsion production, polymer degradation, and degradation of organic compounds (pharmaceutical residues, pesticides, textile dyes, and phenolics). This is the phenomenon of nucleation, growth, and the implosion of vapor or gas-filled cavities in a short period of time (usually milliseconds). It is the transition from the liquid phase to the vapor phase because the local liquid pressure drops to the saturation pressure at a given temperature. At first, cavitation was considered a harmful phenomenon because it caused corrosion, noise, and vibration of equipment, and did great damage to the various surfaces such as marine propellers, pumps, valves, and pipes. These disadvantages can be suppressed or reduced with appropriate measures. The vibration and noise generated during the cavitation process can be solved by better designing pipelines or cavitation devices and using sound insulation materials to reduce noise interference. Besides, when bubbles collapse adjacent to the solid boundary it can result in pitting or erosion of the surface and component failure in the long run.

To effectively reduce the surface damage of pipes, pumps, and valves, high temperature and high-pressure resistant materials should be selected. Meantime, it is vital to control the flow velocity and pressure to avoid water hammer and cavitation onset since it may cause pipe rupture or leakage. Kozak et al. made an attempt to modify the cavitating flow within the Venturi tube by introducing the swirl. In the case of swirl generator presence, cavitation of the boundary layer was less significant, and the volume of the cavitation was surrounded by the liquid water thus reducing the damage of the cavitation erosion to the surfaces of the venturi tube. Simpson and Ranade also reported that the imposition of swirl can move the cavitating region away from solid surfaces toward the device axis, and thus minimize or eliminate the risk of surface erosion.

Therefore, in the process based on venturi or orifice type HC devices, introducing a swirl component may be a good choice to overcome the disadvantages.

Aside from the methods mentioned above, Danlos et al. investigated the effect of different grooved suction-side surfaces of a Venturi-type section on the control of the sheet cavity instability in order to limit erosion and/or noise, and the result demonstrated its feasibility.

When the cavitation bubble bursts, due to the inertia and compressibility of the gas-vapor bubble content, it will generate a huge implosion force, causing local hot spots and releasing large magnitudes of energy. The high temperature (500–15000 K) and pressure (100–5000 atmospheres) are generated as a result of phase change when the bubble implosion happens. This can be explained further from the aspect of the bubble dynamic. In addition to the high temperature and pressure, the collapse of cavitation bubbles can bring various effects which induce numerous physico-chemical changes. Under extreme cavitation conditions, water molecules can be decomposed into a variety of species with a high oxidation potential, including hydroxyl radicals ($\cdot\text{OH}$), $\cdot\text{OOH}$, and H_2O_2 , which are able to react with organic compounds contained in the wastewater.

Besides, the tremendous implosion force caused by the collapse of cavitation bubbles and shear forces can break the molecular bonds of the organic pollutants or make them thermally decomposed and destroy the cell wall of microorganisms, so as to achieve the purpose of degradation of macromolecule organic substance and destroy microorganisms contained in wastewater.

Moreover, when cavities collapse, the micro-jets generate local turbulence and micro-circulation, thereby enhancing the gas-liquid-mass transfer of reactants and increasing the removal of mass transfer resistances in the system. With these cavitation characteristics, it can be utilized as a positive tool to solve or intensify certain processes in specific engineering applications, making it useful and beneficial rather than harmful. The exact cavitation behavior is influenced by the liquid properties (temperature, density, viscosity, and surface tension) and quality, i.e., number of solid particles and amount of dissolved gasses. Both solid particles and dissolved gasses can act as nuclei.

Based on the mode of generation, cavitation can be divided into four categories: optic cavitation (OC), particle cavitation (PC), acoustic cavitation (AC), and hydrodynamic cavitation (HC). In optic cavitation, the light of a pulsed laser converges in a liquid causing the local deposit of energy and then transient bubbles are produced due to optical breakdown. The ultrasonic cavitation process consumes more energy, and the active cavitation zone is also restricted to the vicinity of the transducers, which limits its development and makes it not easy to be amplified.

Therefore, HC (Figure 1) can be employed as an efficient way of process intensification to improve energy and efficiency by enhancing mixing, mass, and heat transfer. Choi et al. employed the process of the HC system combined with Fe (II) to activate the persulfate (PS) to achieve the degradation of pentachlorophenol. Holkar et al. made an overall review of the process intensification (PI) through the cavitation phenomena in which cavitation induced PI, fundamentals, mechanism, advantages, disadvantages or limitations, and case studies have been reported. Panda et al. reviewed the application of HC for a range of processing requirements including HC-assisted extraction, emulsification, food processing, etc. [13].

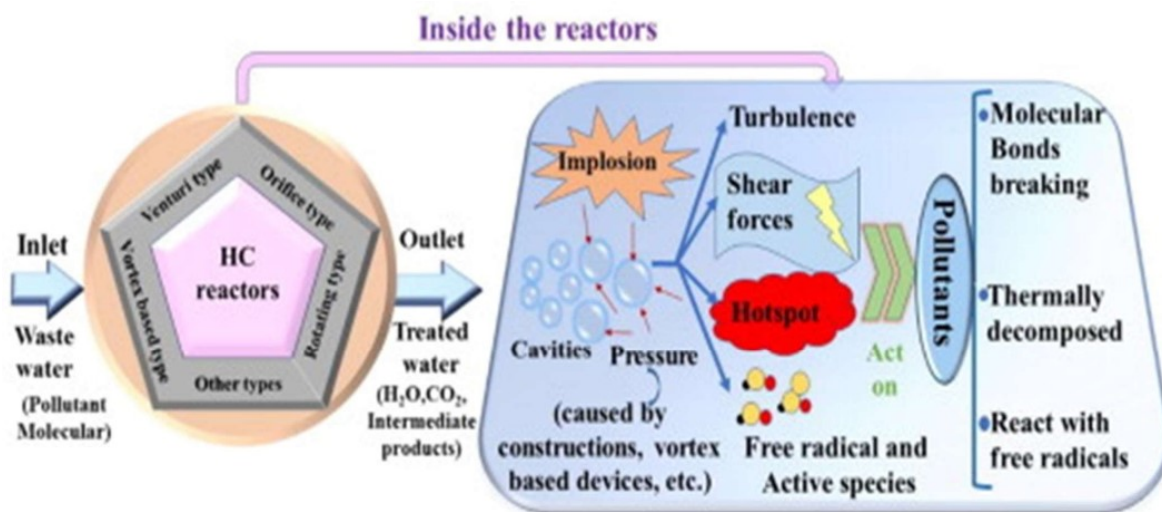


Fig. 1 Hydrodynamic cavitation reactors

The destruction of organic compounds in the wastewater mainly occurs via two pathways: (1) the attack of free radicals takes place in the cavitation bubble, on the interface between the bubble and the surrounding area, and in the bulk solution; (2) pyrolysis occurs inside or near the bubble.

Which of the two mechanisms predominates depends on the properties of the compound and the pattern and intensity of cavitation. Besides, in some cases, the intensity of shockwaves generated by the collapsing cavity can also help to break molecular bonds, especially the complex large molecular weight compounds. The broken-down intermediates are more amenable to be oxidized thus enhancing the rate of oxidation/mineralization of the pollutants. Rajoriya et al. detected the degradation mechanism of Rhodamine 6G driven by the attack of $\cdot\text{OH}$ radicals on the pollutant molecules by using HC with the addition of tert-butanol. Tert-butanol is a type of $\cdot\text{OH}$ scavenger that can neutralize $\cdot\text{OH}$ radicals and produce less reactive tert-butanol radical thus reducing the degradation rate. The results indicated that $\cdot\text{OH}$ radicals were mainly responsible for the decolorization efficiency. HC can be generated by alterations in the flow and pressure, which are usually caused by specific constructions (like venturi tube, nozzle, and orifice plate), vortex-based devices such as vortex diode, or mechanical rotation of the rotating-type devices which are mostly based on the design of a rotor and a stator. The specific constructions function as a throttle valve. When the fluid moves to the throttle, the flow rate or kinetic energy increases at the expense of pressure due to the reduction in the cross-section of the flow passages. Meantime, boundary layer separation and turbulence occur and a substantial amount of energy is lost in the form of a permanent pressure drop due to local fluid turbulence.

In HC, the intensity of cavitation depends on the intensity of turbulence and the number of cavities generated. Cavitation number (C_v), a dimensionless number, is defined to describe the condition of hydrodynamic cavitation. It is given by the following Eq:

$$C_v = \frac{P_2 - P_v}{(1/2)\rho v_0^2} \quad (3)$$

Where P_2 is the fully recovered downstream pressure, P_v is the vapor pressure of the liquid, v_0 is the velocity at the throat of the cavitating device, and ρ is the density of the liquid. C_v decreases with an increase in operating inlet pressure for all the conventional cavitation devices such as orifice or venturi devices since the velocity at the throat of the cavitating device (v_0) increases with the increase of operating inlet pressure. The C_v at which the inception of cavitation occurs is known as cavitation inception number C_{vi} . Studies have shown that the cavitation inception typically occurs at $C_{vi} = 1$ under ideal conditions, and there are significant cavitation effects when the $C_v < 1$.

However, in many cases, due to the existence of small quantities of dissolved gases and suspended particles, cavities can be generated at $C_v > 1$.

Dissolved gases and suspended particles are responsible for the nuclei generation required to initiate cavitation. Besides, the value of C_{vi} is related to many factors, such as the geometry of the flow and the dimension of the constriction.

Yan and Thorpe have reported that the C_{vi} increases approximately linearly with the diameter ratio (β), but for a given orifice, the cavitation inception number does not change with the liquid velocity. Generally, the lower the C_v value, the higher the cavitation intensity, since the number of cavitation events and the generated cavities increase at a smaller C_v , although the intensities of cavity collapse decrease. However, when the C_v decreases, choked cavitation or so-called supercavitation occurs. In this case, the cavities start coalescing with each other and form a cavity cloud as a result of the increased number of cavities, which will reduce collapse pressures [13].

According to Sežun et al., HC can be generally divided into:

- 1) attached cavitation,
- 2) developed or cloud shedding cavitation, and
- 3) supercavitation.

The attached cavitation phenomenon indicates that a large number of vapor bubbles are attached to the constriction surface, forming an attached cloud-like shape. Further, by increasing the flow velocity or decreasing the static pressure in the system, the previously attached cavitation cloud becomes unstable and starts (partly or completely) to shed from the main cavitation structure.

That is the stage of developed or cloud shedding cavitation. Continue to further increases in the flow velocity or reduce the system pressure, the super cavitation occurs. Developed shedding cavitation is the most aggressive one compared to attached cavitation and super cavitation. At the different stages of cavitation, the flow condition will also vary. Saharan et al. analyzed the cavity behavior inside a transparent venturi with the photographic study and confirmed the formation of cavitation inception and choked cavitation with a variation in the inlet pressure.

Depending on the extent of cavitation, the flowing system can have different flow regimes. On the stage of cavitation inception, the single liquid phase first appears as a two-phase bubbly medium in the cavitation zone. Then on the stage of choked cavitation, the flow transitions from two-phase bubbly flow to two-phase annular jet flow, in this case, the central liquid core covered by annular vapor cloud. The formation of choked cavitation is not conducive to producing more energy and active radicals. The C_v for the onset of choked cavitation is called critical C_v .

When using HC for wastewater treatment, the cavitation state should be between the two states of cavitation inception and choked cavitation. Therefore, there usually exists an optimal C_v between C_{vi} and critical C_v . It is essential to find the optimal C_v for a specific reactor to get the maximum cavitation effect whether by numerical simulation or by experiment. It has been reported that the C_v in the range of 0.1–0.2 gives better degradation efficiency. The number of cavitation events and cavities generated increase with decreasing C_v , while the cavity collapse intensities reduce. In addition to C_v , a new parameter, modified cavitation number (C_v') was also applied to characterize the conditions of cavitation by Vichare et al. as shown in equation.

$$C_v' = \frac{C_v}{\text{Total perimeter of holes/perimeter of pipe}} \quad (4)$$

The definition of C_v' considered the influence of the flow geometry on cavitation, and it is of constructive significance when studying the effect of the hole perimeter or hole number. The definition of C_v and C_v' help researchers to analyze and explain the phenomena and results in the HC process to a certain extent.

However, it cannot be ignored that there are some arguments indicating that the cavitation number is not an adequate indication of the inception or intensity of cavitation. For example, Šarc et al. pointed out that large inconsistencies in the reports exist on the cavitation number. It cannot be used as a single parameter that gives the cavitation condition since the geometry, flow velocity, temperature, and gas content all are vital factors influencing the size, dynamics, and aggressiveness of cavitation.

They performed five sets of experiments to inform all the researchers and scientists that using solely the cavitation number as a parameter that defines the cavitation conditions is at least insufficient. Even for the same type of device, small changes to the geometry of the HC devices may cause a significant effect on the cavitation behavior [13].

3 CONCLUSION

By the action of ultrasonic cavitation in aqueous solutions, it is possible to produce a significant concentration of highly reactive OH radicals. By adjusting the ultrasound amplitude, a good selection of physical and chemical parameters and optimization of this advanced oxidation process can be achieved.

This method is very successful for the treatment of wastewater from industrial plants, because harmful pollutants and contaminants after ultrasonic cavitation treatment are not susceptible to genetic mutations. The use of the cavitation procedure for the destruction of chemical and biological substances in wastewater is a method that will be one of the advanced techniques that will have increasing application in wastewater treatment.

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