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RESEARCH OF THE EFFECTS OF VARIOUS GASES ON CAVITATION-BASED REMOVAL OF ORGANIC POLLUTANTS FROM DISTILLERY WASTEWATER

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Досліджено вплив природи газів на процес кавітаційного очищення стічних вод спиртзаводу. Встановлено відносний ряд ефективності впливу природи газів на процес кавітаційного руйнування органічних сполук у стічних водах спиртзаводу. Визначено, що процес руйнування органічних домішок у спиртових стічних водах можна описати кінетичним рівнянням першого порядку. Підтверджено синергічний ефект спільної дії кавітації та азоту при очищенні стічних вод спиртзаводу

Ключові слова: кавітаційне очищення, стічні води, природа газів, хімічне споживання кисню, ступінь руйнування органічних сполук

Исследовано влияние природы газов на процесс кавитационной очистки сточных вод спиртзавода. Установлен относительный ряд эффективности воздействия природы газов на процесс кавитационного разрушения органических соединений в сточных водах спиртзавода. Определено, что процесс разрушения органических примесей в спиртовых сточных водах можно описать, применив кинетическое уравнение первого порядка. Подтверждён синергический эффект совместного действия кавитации и азота при очистке сточных вод спиртзавода

Ключевые слова: кавитационная очистка, сточные воды, природа газов, химическое потребление кислорода, степень разрушения органических соединений

1. Introduction

An important environmental problem of the national economy of Ukraine is wastewater treatment. The methods of economic activity of the country are closely linked to the state and the degree of utilization of water resources and, consequently, the level of economic development. Currently Ukraine tends to irreversible destruction of the environment and of natural resources. Therefore, the use of new technologies in industrial and agricultural wastewater treatment is important.

One of the promising ways to improve the technologies of removing organic contaminants is the use of cavitation in the aqueous medium. Active industrial use of cavitation processes in various industries clearly confirms the high efficiency of this physical phenomenon as a powerful means to accelerate chemical reactions, a mechanism to modify the structures and the properties of aqueous solutions under treatment. This is explained, in particular, by the cavitation-provided powerful energy effect on the treated medium, the inherent processes of phase transformations in the treated liquids, and an acceleration of the chemical reaction.

Given the devastating effects of cavitation in an aqueous medium, the cavitation technology of wastewater treatment can be further improved and developed through directed regulation of the treatment processes by means of using various gases.

2. Literature review and problem statement

The phenomenon of cavitation is used in various industries to intensify mass transfer and hydromechanic processes in the treatment of heterogeneous systems and in the creation of modern energy saving technologies. At present, cavitation technologies are relevant to power engineering, mechanical engineering, as well as chemical and food industries [1]. However, since cavitation affects most of the organic matter contained in wastewater of biotech industries [2, 3], it is used as a stage in wastewater treatment [4].

Cavitation and flotation processes are used to treat the wastewater of meat plants. The use of hydrodynamic cavitation for removal of the typical pollutant such as sodium stearate by means of calcium hydroxide slurry proves that the reaction products emerge on the surface of the liquid and form a dense stable foam. The associated process is flotation; importantly, it occurs without any additional energy supply for the gas dispersion, as opposed to traditional flotation processes. In this case, the gases that cause flotation are emitted during the preliminary stage of cavitation treatment of the aqueous medium. The degree of wastewater treatment due to the use of this method is 85 % [5].

The use of acoustic cavitation in wastewater treatment at the Lviv confectionary Svitoch JSC (Ukraine), where the main components were n-butanol and benzyl alcohol, 1.2–1.7 times accelerated oxidation of the organic impurities [6].

Modern development of cavitation technologies that involves ultrasonic (US) treatment of water systems is reflected in sonochemistry success and, in particular, in the accumulated results on the effect of US on organic [7] and biological objects [8, 9].

When bisphenol A (BPA), which can be found not only in industrial wastewater but also in natural waters, was subjected to ultrasound intensities of 20 W/cm^2, 40 W/cm^2, 60 W/cm^2, and 80 W/cm^2, up to 33.2 %, 44.9 %, 51.1 %, and 55.0 % of it, respectively, was decomposed [10].

Studies [11, 12] show the main role of OH radicals that are formed during cavitation destruction of water in the hydrophilic chemical oxidation of organic pollutants. The volatiles can be burnt in the gas phase in the heat of the bursting cavitation bubbles.

Experiments that involved ultrasonic treatment of domestic wastewater that was contaminated with organic substances proved that cavitation can reduce the COD value by 25–30 % in less than 60 minutes. It was also determined that insoluble compounds were made soluble [13].

Study [14] focuses on the removal of polyvinyl alcohol (PVA) from wastewater by means of ozone/US oxidation. The findings show that ultrasonic frequency and the initial concentration of alcohol have significantly impact the efficiency of treatment. Meanwhile, such characteristics as the reaction time, US power, ozone flow rate, and initial pH have little effects. Maximum removal of organic compounds in terms of COD is achieved under the following conditions: the initial concentration of PVA of 100 mg/dm^3, initial pH – 9, ozone dose – 4 g/h, US power – 320 W, US frequency – 40 kHz, and reaction time – 20 min. In these conditions, the efficiencies of COD and PVA removal are 86.4 % and 99.3 %, respectively. The use of O^3/US improves the COD removal efficiency by 19.4 % compared to the use of ozone alone under similar optimal conditions.

Parameters such as US power, dissolved gas, pH, and initial concentration of ibuprofen (IBP) were studied in [15], in ultrasonic treatment (300 kHz). US removed 30 to 98 % of IBP within 30 minutes. Under the influence of air and oxygen, the decomposition rate of IBP was 4 μmol l^{-1} min^{-1}, which is higher than with the use of argon. After 120 minutes of treatment, IBP was completely removed to the value of biological oxygen demand (BOD) of 8 %. At the same time, the biodegradability (BOD_5/COD) reached 0.36. Thus, ultrasound provides complete IBP removal via transforming IBP into biodegradable products that are removable in subsequent biological treatment.

A strengthened cavitation effect and the simultaneous use of gas and ultrasound allowed achieving a high rate of destroyed cells faster than when each agent acted alone [16, 17]. This indicates the feasibility of gas cavitation processes in water disinfection and proves an intensive destruction of microbes affected jointly by gas and ultrasound. When gas bubbles in the cavitation zone, the slurry is effectively mixed in the ultrasound field, the flow becomes turbulent, and there appears a synergistic effect as well as further signs of cavitation. The presence of a synergistic effect reduces energy costs in the cavitation treatment of aqueous solutions because of its lower duration compared to the uses of gas or ultrasound separately. Such joint action of the factors can increase the effectiveness of treatment processes and opens up the prospects for practical application of US in the presence of gases of various nature.

Supplying various gases to the cavitation zone can influence the chemical and acoustic removal of radicals, the process mechanism, and the course of US chemical processes. However, certain regularities of gas effects on the kinetics of chemical reactions in the cavitation field are still insufficiently investigated, theoretically justified, and, thereby, necessitate further research in the area.

3. Research purpose and objectives

The aim of the paper is to study the effect of various gases on the cavitation removal of organic pollutants from distillery wastewater.

To achieve this goal, we set the following tasks:

– to measure the effects of nitrogen, oxygen, and a mixture of nitrogen and oxygen in the ratio of 1:1 on the distillery wastewater treatment in cavitation conditions and without them;

– to determine how the kinetic regularities and peculiarities of ultrasonic cavitation affect the distillery wastewater treatment depending on the nature of the bubbled gas.

4. Materials and methods of studying gas effects on the cavitation wastewater treatment

4. 1. Researched materials and experimental equipment

The effect of the nature of the bubbled gas on the cavitation treatment of wastewater was studied with the use of such gases as nitrogen, oxygen, and a mixture of nitrogen and oxygen in the ratio of 1:1. Potassium dichromate $K_2Cr_2O_7$, ammonium iron(II) sulfate (or Mohr's salt), concentrated sulfuric acid, N-phenylanthranilic acid solution, and distilled water served as excipients.

The influence of cavitation on removing organic substances from the wastewater of a distillery plant was studied with the use of a low-frequency generator USDL-2T (Ukraine) (its rated power is 90 W). Ultrasonic oscillation with a frequency of 22 kHz was transmitted with the help of a magnetostriction emitter immersed in the water under study with a known initial value of chemical oxygen demand (COD).

4. 2. Methods to determine the characteristics of wastewater quality

Wastewater is usually a mixture of organic and inorganic compounds. The methods used to assess the quality of wastewater are divided into methods for measuring the total content of organic matter in wastewater and methods for measuring the contents of certain organic compounds. The basic parameters of water quality include biochemical oxygen demand (BOD), chemical oxygen demand (COD), and total organic carbon (TOC).

In international practice, there are numerous BOD standards such as SFS-EN 1899-1 (SFS-EN 1899-1), APHA Standard 5210 B (APHA, 1998), and OECD 301 F-guide (OECD, 1992). For wastewater samples, the standard measurement is five days (BOD_5) at 20 °C, but there can be used a different time and a different temperature. In Finland, for example, a typical measurement time is seven days (BOD_7) (Karttunen, 2003). The COD value is determined in accordance with APHA 5220 "The chemical oxygen demand (COD)" [18].

In the study of distillery wastewater, the main parameter to assess the quality of wastewater was the COD value. Alongside COD, there were determined effective rate constants for the organic compounds destruction (k) and the degree of organic compounds destruction (D).

The bichromatic method was used to determine COD, i. e. the amount of oxygen (mg) that is required for the chemical oxidation of all organic and inorganic agents in 1 dm^3 of water. Initial COD_0 values were in the range of 500–1000 mgO_2/dm^3. The output values of COD/COD_0 were used to represent the experimental data.

The effective rate constant for the organic compounds destruction was determined with the use of the first-order kinetic equation.

The degree of the organic compounds destruction (in %) was calculated by the following formula:

$$D = 100 - \frac{COD \cdot 100}{COD_0}, \quad (1)$$

where COD_0 is the initial value of COD, mgO_2/dm^3, and COD is the current COD value at a certain moment of time τ, mgO_2/dm^3.

5. Research findings on wastewater quality

The determined COD values that depend on the time of the distillery wastewater cavitation in the presence of nitrogen are shown in Fig. 1. It should be noted that the highest cleansing effect is achieved when US and gas are used jointly. Already after the first hour of US treatment in the presence of nitrogen, there was an evident increase in the cleansing action compared with separate actions of either ultrasound or nitrogen.

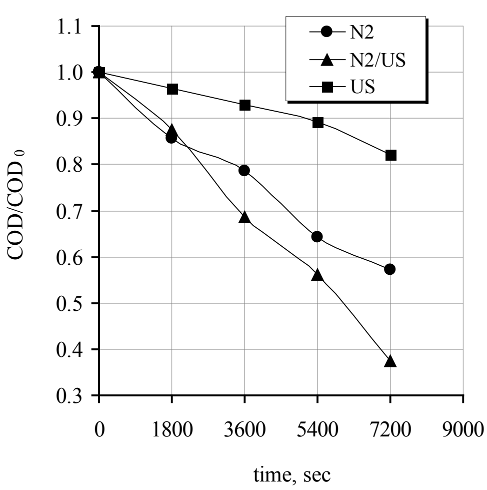

Fig. 1. Dependence of COD/COD_0 of distillery wastewater on the time of treatment in the presence of nitrogen

In two hours, the ultrasonic treatment of wastewater in the presence of nitrogen reduces the content of organic matter to 199 mgO_2/dm^3, which is 2.67 times lower compared to the initial value of COD_0=532 mgO_2/dm^3. Nitrogen alone reduces the COD 1.74 times compared to COD_0=465 mgO_2/dm^3. Ultrasound alone reduces the COD by 133 mgO_2/dm^3, which is 1.2 times lower compared to COD_0=798 mgO_2/dm^3. Ultrasound alone is by 56 % less effective than if it is used jointly with a gas. The cleansing effect of nitrogen alone and its effect together with ultrasound in the first 30 minutes of reaction are almost identical, but an hour later there is observed a significant destruction of organic impurities in the ultrasound field.

The research findings on the effect of oxygen on the cavitation treatment of distillery wastewater show that the most effective removal of organic impurities is achieved in the joint action of ultrasound and oxygen. Either the suply of oxygen alone to the reaction zone or the use of ultrasound alone has a much lower effect. Already after the first hour, the action of ultrasound in the presence of oxygen manifests an increase in the cleansing action compared to the effect of ultrasound alone. As for oxygen, it is effective only in combination with ultrasound, whereas US alone is 32 % less effective compared to the action of O_2/US.

The results of further studies that used a joint bubbling of nitrogen and oxygen in the ratio of 1:1 both in the ultrasound field and without it are presented in Fig. 2. As well as in previous experiments, the most appropriate is the joint use of the gas mixture and ultrasound.

When the gas mixture alone is passed through the water, the COD is reduced to 399 mgO_2/dm^3, which is 1.33 times lower compared to the initial value of COD_0=532 mgO_2/dm^3. The mixture of gases gives the result close to the action of ultrasound that reduces the COD 1.2 times.

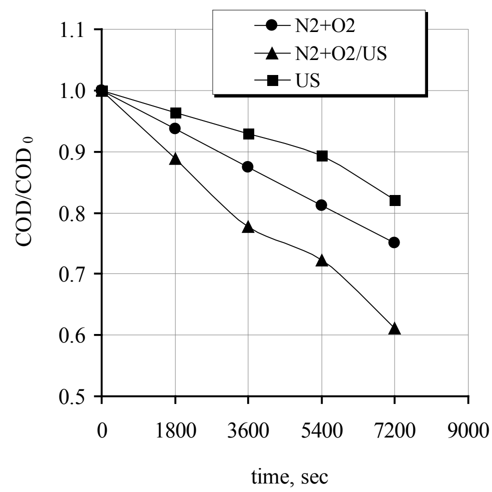

Fig. 2. Dependence of COD/COD_0 of distillery wastewater on the time of treatment in the presence of the mixture of nitrogen and oxygen in the ratio of 1:1

In two hours, the ultrasonic treatment of wastewater in the presence of the gas mixture reduces organic matter to 352 mgO_2/dm^3, which is 1.63 times lower compared to the initial value of COD_0=576 mgO_2/dm^3. The use of ultrasound alone is by 26 % less effective than its joint use with the mixture of gases.

Analysis of the results of experiments reveals that the destruction of organic compounds occurs in all cases; the difference is only in the effectiveness of the organic impurities destruction that depends on the bubbled gas (Fig. 3).

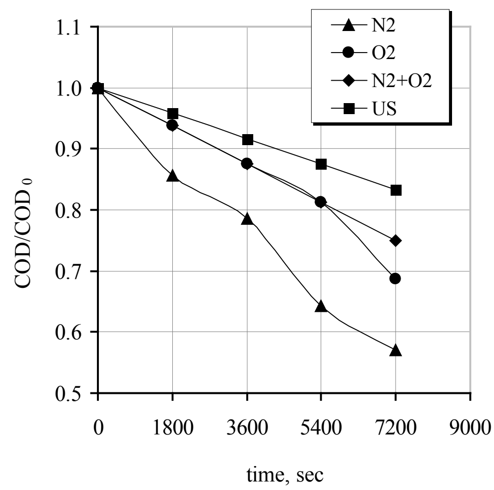

Fig. 3. Dependence of COD/COD_0 of distillery wastewater on the time of treatment with various gases

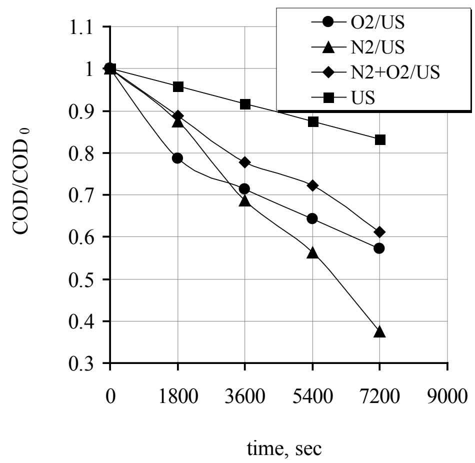

Fig. 4. Ultrasound-chemical dependence of COD/COD_0 of distillery wastewater on the time of treatment with various gases

In the study of the impact of the nature of gas on the destruction of organic compounds in wastewater (Fig. 3), we should particularly emphasize the effect of nitrogen. Within the two-hour treatment, the highest cleansing effect is achieved due to the use of nitrogen. It was experimentally proved that the action of nitrogen reduces the COD 1.74 times compared to the initial value of COD_0=465 mg/dm^3, oxygen reduces the COD 1.45 times (COD_0=532 mg/dm^3), and a mixture of gases – 1.33 times (COD_0=532 mg/dm^3). The least effect in removing organic contaminants from wastewater was achieved when ultrasound was used alone – the COD became 1.2 times reduced (COD_0=798 mg/dm^3). The use of nitrogen in the wastewater treatment had a pronounced cleansing effect already during the first few minutes of bubbling, which is not observed in the presense of other gases and especially ultrasound used alone. As shown in Fig. 4, the action of oxygen and gas mixtures during up to 90 minutes of the experiment result in almost the same oxidation. 90 minutes of the reaction result in a significant COD decrease. In the presence of oxygen, the COD decreased by 66 mg/dm^3 compared to a mixture of gases – 33 mg/dm^3. Under these conditions, the effective rate constant for the organic compounds oxidation is $0.5 \cdot 10^{-4}$ sec^{-1} in the presence of oxygen vs. $0.4 \cdot 10^{-4}$ sec^{-1} in the presence of the mixture of nitrogen and oxygen (Table 1).

Table 1

Summary of the effective rate constants for the destruction of organic compounds in distillery wastewater

Bubbled gas	$k \cdot 10^4$, s^{-1}	
	without US	with US
N_2	0.7±0.06	1.2±0.07
O_2	0.5±0.04	0.8±0.06
N_2+O_2	0.4±0.02	0.7±0.06
US	0.2±0.02	–

When cavitation treatment of wastewater was held in the presence of nitrogen, oxygen, and the mixture of nitrogen and oxygen in the ratio of 1:1 (Fig. 4), there was observed a COD reduction during the entire process for all of the gases under study.

During the first 30 minutes of ultrasonic treatment of the wastewater, the highest effectiveness is exhibited by oxygen, after which the curve smoothly descends. As shown in Fig. 5, bubbling of the gas mixture (of nitrogen and oxygen) and oxygen alone results in almost the same final values of COD; the difference is only in the rate of organic compounds destruction. But when the treatment time is increased, the highest effect is achieved due to the bubbling of nitrogen in cavitation conditions. The value of the effective rate constant for the organic substances oxidation is $1.2 \cdot 10^{-4}$ s^{-1}, and the degree of purification reaches 63 %. The lowest result is observed under the action of ultrasound alone: the COD reduces to 665 mgO_2/dm^3, which is 1.2 times lower compared to the initial value of COD_0=798 mgO_2/dm^3.

The reaction order and the effective rate constant for the organic compounds destruction are determined with the use of the first-order kinetic equation. Straightening in the coordinates $\ln(COD/COD_0) - \tau$ (Fig. 5, 6) confirms that the process of removing organic contaminants in the distillery wastewater treatment can be described by the the first-order kinetic equation.

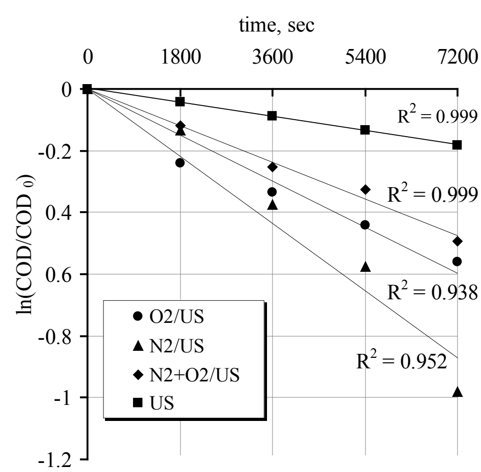

Fig. 5. Semilogarithmic dependence $\ln(COD/COD_0)$ of distillery wastewater on the time of treatment with various gases

The effective rate constants for the organic compounds destruction in distillery wastewater are determined analytically with the use of the integral method by the following equation:

$$\ln\frac{COD}{COD_0} = -k\tau, \quad (2)$$

where k is the effective rate constant, s^{-1}, and τ is the time of destruction of organic compounds in wastewater, s.

High correlation coefficients shown in Fig. 5, 6 confirm the adequacy of the model. The error of calculation of the rate constants for the destruction of organic compounds in wastewater was determined by three parallel experiments and did not exceed 8 %.

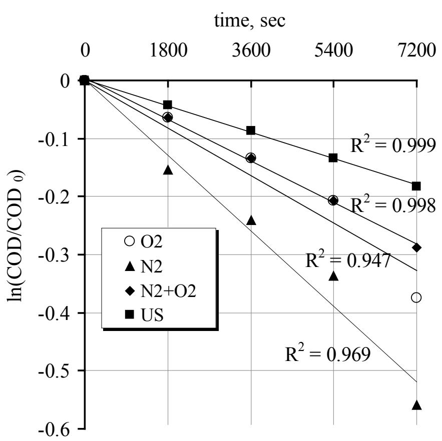

Fig. 6. Semilogarithmic dependence $\ln(COD/COD_0)$ of distillery wastewater on the time of treatment with various gases

Table 1 shows the values of the effective rate constants for the destruction of organic compounds in distillery wastewater with their confidence intervals.

As shown in Table 1, in cavitation conditions, the use of nitrogen results in the highest value of the effective rate constant – $1.2\cdot10^{-4}$ s^{-1}.

Oxygen alone and the mixture of nitrogen and oxygen in the ultrasound field have almost identical values of the constants – $0.8\cdot10^{-4}$ s^{-1} and $0.7\cdot10^{-4}$ s^{-1}, respectively, but this is almost 4 times higher compared to ultrasound alone – 0.2 10^{-4} s^{-1}. The effective rate constant for the destruction of organic matter confirms the high efficiency of nitrogen combined with US. This value is 1.5 times higher compared to oxygen, and 1.71 times higher compared to the gas mixture. During bubbling the gases alone (without ultrasound), in the nitrogen-dominated atmosphere the effective constant is 0.7 10^{-4} s^{-1}, for oxygen it is 0.5 10^{-4} s^{-1}, and for the mixture of gases – 0.4 10^{-4} s^{-1}.

Table 2

Summary of the degrees of distillery wastewater treatment in cavitation conditions in the presence of various gases

Bubbled gas	The degree of destruction D, %	
	without US	with US
N_2	42	63
O_2	31	43
N_2+O_2	25	39
US	–	17

It is also found that the effective rate constant for wastewater treatment at the bubbling of nitrogen in the ultrasound field is higher than the sum of effective rate constants for the same agents used alone. This poves the synergy of action of US and gas on the removal of organic pollutants from the wastewater.

$$k_{(N2/US)} > k_{(US)} + k_{(N2)};\ (1.2\cdot10^{-4}\ s^{-1} > 0.2\cdot10^{-4}\ s^{-1} + 0.7\cdot10^{-4}\ s^{-1}).$$

The nature of the gas predetermines varying degrees of destruction of organic impurities. The highest degree of destruction of organic matter is reached during ultrasonic treatment in the presence of nitrogen (63 %) (Table 2). After a two-hour ultrasonic treatment and oxygen bubbling, the achieved effect is lower – 42 %, whereas for the gas mixture it is 38.8 %. The lowest effect – 17 % – is observed when ultrasound is used alone.

The obtained data reveal a series of effects of the gas nature on the destruction rate of organic compounds (that are contained in the distillery wastewater) during the cavitation treatment:

$$N_2 > O_2 > N_2 + O_2.$$

The research confirms the expediency of the joint action of gases and ultrasound in the distillery wastewater treatment, with the highest efficiency in cavitation conditions demonstrated by nitrogen.

6. Discussion of the findings on the effects of various gases on cavitation in distillery wastewater treatment

The study of the impact of nitrogen on the process of the distillery wastewater treatment that is presented in Fig. 1 reveals an abrupt and stable COD decrease during the whole process of cavitation. However, the highest cleansing effect is achieved when the gas is bubbled in the cavitation zone. The joint use of ultrasound and nitrogen is by 46 % more effective than the the use of ultrasound alone.

In addition to nitrogen supplied to the cavitation zone, the nature of oxygen and the mixture of nitrogen and oxygen in the ratio of 1:1 also contribute to initiating the treatment processes (Fig. 2). The obtained results prove the effectiveness of ultrasound in the treatment of distillery wastewater. The bubbling gas in the ultrasound field reduces the COD, thereby, increasing the degree of wastewater treatment, the value of which depends on the nature of the bubbled gas (Table 2).

Comparative analysis of the effects of various gases on the wastewater treatment (Fig. 3) shows that the joint use of nitrogen and oxygen in the ratio of 1:1 and oxygen alone have somewhat similar effects. The most effective was the impact of nitrogen, even compared to the effect of ultrasound.

The presented data (Fig. 4) show that in the process of water treatment, the most effective is the joint effect of ultrasound and gas that are used simultaneously. In the cavitation field, the oxidative effect of oxygen is significantly higher in comparison with the use of oxygen alone. A similar pattern is observed for the mixture of nitrogen and oxygen in the ratio of 1:1. Thus, the research data prove an important role of the nature of the gas with which water is saturated in ultrasonic treatment. The dissolved gas exhibits a dual effect: on the one hand, it ensures a surplus of primordia for the formation of cavitation bubbles. The gas improves the conditions required for the destruction of bubbles since it raises the temperature at their compression. This increases pressure in the bubbles, reduces the rate of their cavitation, and increases the entire cleansing effect.

One of the main features of ultrasonic chemical reactions is the fact that they occur in water saturated with a particular gas. Water is not only the most favorable medium to form cavitation but also a medium that provides the best opportunity of better electronic breakdown of the cavity. The reaction participation by water molecules that are sources of H and OH radicals is reduced to initiating most chemical reactions. Such conditions are likely to excite molecules and atoms of various gases (N_2, O_2, N_2+O_2, etc.). The radicals are partially recombined; the composition of the end radical and molecular products of water decomposition in the ultrasound field depends on the nature of the dissolved gas. Moreover, the water molecules' excitation and their dissociation rate are largely dependent on the presence of excited gases in the cavities.

Cavitation cavities are penetrable to water vapors, dissolved gases, and inpenetrable to ions or molecules of nonvolatile solutes. In the bubble collapsing, any of the present gases actively participates in the transfer of excitation energy and recharging. The effect of the ultrasound field on the substances that penetrate into the cavity is direct and obvious, whereas the effects of active gases such as O_2 and N_2 in the cavity are twofold:

– firstly, O_2 participates in the reactions of radical transformations:

$$H^{\bullet}+O_2 \rightarrow HO_2$$

and N_2 – in ultrasonic-chemical gas reactions that are targeted at fixing nitrogen:

$$N_2+O_2 -))) \rightarrow NO_2;$$

– secondly, reactive gases penetrating in the cavity participate alongside the noble gases in the transfer of electronic excitation energy to water molecules, as well as in the recharge process [19, 20].

It is found that the joint use of ultrasound and gas increases the destruction rate of organic contaminants 1.7–3.5 times (Fig. 5, 6) since passing gas through ultrasonically treated wastewater enhances the destruction of organic substances. Gas bubbles act as additional promordia of cavitation that intensify the formation of such radicals as H and OH. These radicals act as strong oxidants, which is proved by the found synergistic effect when gas and ultrasound are jointly used in cavitation. The nature of nitrogen allows reaching the highest effective rate constant for the destruction of organic compounds (Table 1) and, consequently, the highest degree of wastewater treatment (Table 2).

7. Conclusions

1. It is shown that the joint cleansing effect of bubbling gases in the cavitation field is by 22–46 % higher compared to the effect of ultrasound alone and by 12–25 % higher than the effect of the gases alone. This confirms the revealed synergistic effect of the joint use of gas and ultrasound in the distillery wastewater treatment.

2. The research has revealed the relative series of effects of the nature of gases on the process of cavitation treatment of distillery wastewater. It is determined that the effective rate constant for the destruction of organic compounds depends on the nature of the bubbled gas. The highest cleansing effect on the cavitation destruction of organic compounds is demonstrated by nitrogen. Its use in the cavitation field can improve the degree of the wastewater treatment by 46 % compared to the effect of ultrasound alone.

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