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SONOCHEMICAL METHODS OF REMOVING THE ORGANIC IMPURITIES FROM WATER

METODY SONOCHEMICZNE USUWANIA ZANIECZYSZCZEŃ ORGANICZNYCH Z WODY

Abstract: Surface water is characterized by changeability of composition/content and organic intensity of both natural and anthropogenic impurities. As a result of chemical changes developed in the process of oxidation and disinfection, impurities and organic admixtures may become harbingers of damaging by-products of these processes (OBP/DBP). Thus, removing organic impurities from water is a crucial problem in water technology, which predominates in the research subject matter. The decrease of water organic contamination can be obtained by the following methods: coagulation, adsorption, oxidation and membrane processes. The use of different solutions in this field has been researched recently as well. The present research is aimed at so-called 'hybrid methods' combining chemical methods with physical factors. For instance, the processes of profound oxidation, in which ultraviolet or ultrasounds are used as hydroxyl radical initiators.

The literature based research presented in the first part of the article indicates that the unconventional ultrasound method was described on the basis of the effects obtained with the use of prepared water (most frequently with commercial humic acids preparation). Due to the composition of humic substances in surface water (where fulvic acids are predominant), the verification of these effects in natural water environment is justified. In the following part of the article we presented the research results concerning the effects of the use of ultrasound field with the high intensity and constant frequency of 24 kHz. The substrate for the research was surface water. The laboratory tests were conducted at changing sonification time and vibration amplitude. Changes in water organic impurity were controlled mainly by the TOC index analyses. What was researched as well, was the influence of water pH on the removal of organic impurities measured by the TOC index. In order to characterize it the experiments were conducted at natural water pH, alkaline and acid reaction. At the beginning of the experiment and after the use of the researched sonochemical method we marked the chosen parameters: TOC, DOC, oxygen consumption, UV₂₅₄, and pH. On the basis of these parameters we evaluated the effects of the process depending on the changing ultrasound parameters.

Keywords: water treatment, organic impurities, ultrasounds

Nowadays, water is exposed to rapid contamination, and it is presently hard to find water of natural physicochemical composition. It is obvious that contaminants find their way to surface water as a result of human activity, but the concentration of natural components is determined by natural conditions. However, in water subjected to

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treatment any undesirable substance that is to be removed in the technological process is considered a contaminant, regardless its origin. The reduction of the organic contamination of water is achieved by the following methods: coagulation, adsorption, oxidation, and membrane processes [1].

Humus substances (HS) are present in almost all natural waters and may account for 60–80 % of the overall mass of *natural organic matter* (NOM) occurring in water and bottoms [2]. Their contents depend on the type of soil, from which they have been washed out, water contact time, the chemical composition and pH of water. The predominant substances dissolved in surface water are fulvic acids, constituting up to 87 % HS, their sodium or/and potassium salts; *humic acids* (HA) (2 % HS) and hymatomelanic acids are less frequent. In ground water, HA are prevalent. HS present in water can be a source of an undesirable smell and impart a colour (ranging from light yellow to dark brown) to the water, being characteristic of a specific group of acids. In addition, HS form complex compounds with heavy metals, iron, aluminium, and pesticides occurring in natural water, to maintain these pollutants in a dissolved form. Previously, only the water colour and smell justified the removal of those compounds from water being treated, as humus substances naturally occurring in quantities below 100 g/m³ are not hazardous to health. However, organic compounds present in water subjected to oxidation and disinfection processes are responsible for the formation of undesirable halogenated hydrocarbons referred to as *oxidation/disinfection by-products* (OBP/DBP). They exhibit adverse health properties, and even mutagenic or carcinogenic effects [3].

In Poland, gaseous chlorine is still most frequently used in disinfection processes; chlorine dioxide and ozone are less frequently used, and ultraviolet radiation is employed sporadically [4]. The chlorination of water (Cl₂ treatment) leads to a reaction of substitution in organic matter molecules, as a result of which toxic chlorinated organic compounds, such as trihalomethanes (with their precursors being mainly HS), halogenoacetic acids, halogenoketones, or trichlorophenols, form. To reduce the risk of the chlorination reaction occurring, gaseous chlorine is substituted with chlorine dioxide with higher oxidation potential. The by-products of oxidation with ClO₂ may include chlorates(V), and chlorites(III), which are also categorized into DBPs. The application of ozone intended to eliminate the formation of chlorinated organic compounds causes the oxidation of inorganic compounds and oxidation of organic compounds into carbon dioxide and water. However, sometimes it results in harmful intermediate products, such as ketones, aldehydes, or carboxylic acids. Pre-oxidation processes result in a reduction of water colour, but this is not always indicative of a sufficient removal of organic matter [5]. Therefore, new possibilities for removing biodegradation-resistant organic substances are being searched for.

The need for minimizing the amount of reagents used in the water treatment process and reducing the *total organic carbon* (TOC) content to the lowest possible level has initiated searches for organic contaminant removal methods that would be more effective, while not leading to the formation of OBP/DBP and reducing the risk to the natural environment. Hence, AOP (*Advanced Oxidation Process*) methods are increasingly popular and of great interest. These are methods that combine the chemical

oxidation process with physical factors. The following can be employed in various combinations: ultrasounds, ultraviolet radiation, and additions of catalysts (TiO_2 , MnO_2 , Fe^{2+} and Fe^{3+}). The application of these methods results in the formation of hydroxyl radicals (OH^\cdot) of a very high oxidation-reduction potential of 2.8 V. Moreover, hydroxyl radicals are characterized by the ability to non-selectively react with many organic compounds, whereby they are sufficiently effective in removal of organic compounds by transforming them into carbon dioxide, water and inorganic compounds. It is important to note that even with an incomplete oxidation of organic matter simpler forms with smaller molecular masses are obtained, which are susceptible to biodegradation [6, 7].

Literature review

Owing to the ultrasonic cavitation phenomenon, the *ultrasonic* (US) field can also independently initiate the formation of hydroxyl radicals, hydrogen dioxide, and ozone. Free radicals generated become the main source of so-called sonochemical reactions. Their intensity, with additional combination of ultrasounds (UD) with chemical reagents or a gaseous phase (air, argon) introduced, may be increased even further. By reacting with the molecules of various substances (dissolved or suspended in water), hydroxyl radicals cause the chemical decomposition, reduction or destabilization of those molecules. The effect of sonochemical methods on the degradation of numerous organic compounds has been confirmed by some studies; however, model substances, such as carbon tetrachloride (CCl_4) or phenol were mostly used in those studies. Studies on the application of an unconventional ultrasonic method for the removal of organic water admixtures are described mainly based on the effects obtained for water solutions prepared using commercially available humic acid (HA) preparations. However, HAs do not represent the surface water environment, where *fulvic* acids (FA) are predominant. Example results of those studies are presented below.

Naddeo et al [8] studies the effectiveness of ultrasonic removal of natural organic matter (NOM). The studies showed that the degree of NOM removal was dependent on the duration and intensity of sonification. Solutions, each of a volume of 300 cm^3 , were prepared using a commercial HA preparation supplied by Aldrich. Sonification was conducted using a VCX-750 disintegrator by Sonics & Materials, equipped with a 1.3 cm-diameter sonotrode (with a frequency of 20 kHz, and a field intensity from to 42 W/cm^2). The NOM content was measured as TOC and absorbance (at $\lambda = 220\text{--}620 \text{ nm}$). The authors noted a reduction in HA concentration (measured as TOC) at a level from 24.5 % to 34.9 % after 20 minutes of sonification and at a field intensity of 42 W/cm^2 . The results of absorbance measurements indicated that a reduction in organic matter contents had resulted, whereas the values of the TOC index did not show the same effectiveness. The increase in the absorption of UV_{254} radiation and the increase in the turbidity of solutions subjected to sonification are explained by the authors by an aggregation of split fragments of humic acids. They also point out that the TOC measurement is not affected by these phenomena, as intermediate forms of organic compounds are also subjected to analysis. Owing to this, the determination of the TOC index is more universal.

Y.-S. Ma and J.-G. Lin [9] investigated the effect of different methods using ultrasounds on HA removal from water. For this purpose, they compared the effectiveness of three systems: ultrasounds, O₂ and ultrasounds/O₂. An ultrasonic generator with the following parameters was employed: frequency, 20 kHz; power, up to 160 W; and intensity, 126.5 W/cm². The parameters representing the effectiveness of a specific system were: oxidation-reduction potential, BOD₅ (*Biochemical Oxygen Demand, 5-day*), and TOC. The substrate for the tests were solutions prepared from a HA preparation (HO161, supplied by Tokyo Chemicals) and deionized water. The initial TOC concentration was 18.6 ± 0.4 mgC/dm³. The coupled ultrasound/O₂ process turned out to be the most effective in HA mineralization. After 120 minutes of sonification the TOC was reduced by 52.6 %. Not combined, these methods only showed a TOC reduction in the case of US (26.5 %) and O₂ (27.4 %) alone. The authors showed that the hybrid process (US/O₂) increases TOC removability and reduces chlorine demand.

Chemat et al [10] state that an alternative for conventional oxidation methods is the application of a high-intensity (>10 W/cm²) US field in combination with the use of a commercial oxidizer, that is H₂O₂. In the researchers' assumption, this system was supposed to provide an increase in the degradation and mineralization of organic compounds. A field of an intensity of 20 kHz and a power of 50 W (20 W in the centre) was used. The determinations of TOC and UV₂₅₄ absorbance, both correlating with the HS contents, were made. The reduction in the value of absorbance indicated a complete removal of synthetic HAs after 60 minutes of sonification, which was not confirmed by the TOC analysis (a removal of a mere 40 %). The authors explain this by the accuracy of the TOC assay that detects also intermediate products. They claim that after 180 minutes of sonification the reduction of TOC was at a level of 90 %. At the subsequent stage of experiment, the effectiveness of an oxidizer aided by a magnetic stirrer (50 rpm) and an oxidizer aided by ultrasounds was compared. The mineralization of organic compounds in the first model was 25 %, while for the same contact time of the oxidizer, but US aided, a mineralization degree of 90 % was achieved. The aim of the investigation was also to determine the effect of pH and H₂O₂ concentration on the HA degradation. The best results (50 % relative to absorbance at λ = 254 nm) were obtained for pH = 3. The analysis of the effect of oxidizer dose (0, 50, 100, 200, 300, 500) mg/dm³ for a solution of 100 mg/dm³ HA showed that the best process effectiveness had been achieved for an H₂O₂ concentration of 200 mg/dm³.

Results of studies on the removal of THMs (Trihalomethanes), compounds formed as a result of the chlorination of *eg* humus substances, from water solutions are also known. In an experiment described by H. Shemer and N. Narkis [11], an ultrasonic processor (20 kHz, field intensity 3.75 W/cm²) was employed. The authors made a summary of THM removal effectiveness as a function of sonification time. Within 180 minutes of US field operation, 100 % CHCl₃, 80 % CHBr₃ and 60 % CHI₃ were removed sonochemically.

Thus, the literature review indicates that the effectiveness of the unconventional ultrasonic method described in the hitherto existing studies on prepared waters not always can be taken into account in the water treatment technology. From the point of

view of the composition of humus substances, particularly those contained in surface water (where fulvic acids are predominant), it is justifiable to verify these effects in a natural water environment.

Experimental

The aim of the experimental studies carried out was to assess the effectiveness of reducing the contents of organic and inorganic compounds in surface water by sonochemical methods with the use of a high-intensity ultrasonic field. Natural water was taken from the water reservoir at Poraj near Czestochowa, Poland. Samples, each of a volume of 500 cm³, were subjected to sonification. An Hielscher UP400S disintegrator with the following parameters was used:

- vibration frequency: 24 kHz,
- effective power: 400 W (300 W in water),
- H22 sonotrode diameter: 22 mm ($S = 3.8 \text{ cm}^2$),
- maximum ultrasonic field intensity: 85 W/cm² (for the selected sonotrode and a maximum amplitude of $A = 60 \text{ }\mu\text{m}$).

The effect of the vibration amplitude and sonification time, as basic ultrasound parameters determining the level of field intensity and energy input, were assessed. To this end, the following combinations of ultrasonic parameters was examined: a constant vibration amplitude and the sonification time variable in the range from 2 to 8 minutes, and a constant sonification time with a different vibration amplitude of 18, 36, and 54 μm , respectively. As each of the individual humus substance fractions dissolves in its specific environment, the experiment was conducted at the pH of natural water (pH = 7.98), at an alkaline reaction (pH ~ 9), and at an acid reaction (pH ~ 3). The reaction of water was changed by adding solutions of HCl (1:1) and 30 % NaOH to it. The examinations of water samples at the above-mentioned pH values were also aimed at determining the effectiveness of the oxidizing effect of radicals on organic substances, depending on the reaction of a sample. At the beginning of the experiment and upon the application of the methods under examination, the assays and measurements of TOC, DOC (Dissolved Organic Carbon), oxygen consumption, UV₂₅₄ and pH were made. On their basis, the effects of a process were assessed, depending on the variable ultrasonic parameters. The basic assay used for the assessment of change in the contents of organic compounds in the water was the TOC index. The determination of TOC and DOC were made according to the standard PN-EN 1484:1999 using a Multi N/C 2000 analyzer (the DOC index was determined after the sample had been filtered through a 0.45 μm mesh filter). Absorbance was measured with an Helios α Spectrometer in 5 cm-long optical cells. Oxygen consumption (permanganate value) was determined according to the standard PN-EN ISO 8467:2001.

Analysis of investigation results

The examined water was characterized by the following parameters: TOC = 10.27–14.69 mgC/dm³; DOC = 8.01–11.97 mgC/dm³; oxygen consumption: 9.79–10.4 mgO₂/dm³;

pH = 0.92–8.55; turbidity: 8.31–4.9 NTU. These indicate an increased organic contamination of water.

Two combinations of ultrasonic field parameters were applied. The first of them consisted in the variation of the amplitude with a selected fixed sonification time. The effect of ultrasonic parameters visible in the examination results (Table 1) indicates that the vibration amplitude has a major importance for the process effects. Increasing the vibration amplitude enables an increase in ultrasonic field intensity to be obtained, which for $A = 54 \mu\text{m}$ amounted to approx. 75 W/cm^2 . For the highest amplitude value and a sonification time of 3 minutes, the most favourable reduction of the TOC index (by 6.64 mg C/dm^3) was noted.

Table 1

Effect of amplitude and sonification time
on the selected indicators of water (natural water pH) [12]

Index	Before UD* natural water pH	After UD					
		3 min			18 μm		
		18 μm	36 μm	54 μm	2 min	5 min	8 min
pH	7.92	7.78	7.74	7.71	7.8	7.73	7.7
TOC [mgC/dm^3]	14.69	11.14	10.11	8.05	9.56	10.62	11.02
DOC [mgC/dm^3]	11.97	—	—	7.88	—	—	10.72
Oxygen consumption [mgO_2/dm^3]	10.4	9.15	8.32	7.71	8.74	8.32	7.9
Absorbance UV_{254}	1.01	1.19	1.08	1.02	1.1	1.09	1.31

* UD – ultrasounds.

For these parameters, the effectiveness of TOC reduction was 45 % (Fig. 1A), whereas for DOC it was lower, amounting to 35 %. As a result of the sonification process, dissolved fractions, accounting for more than 80 % TOC for crude water,

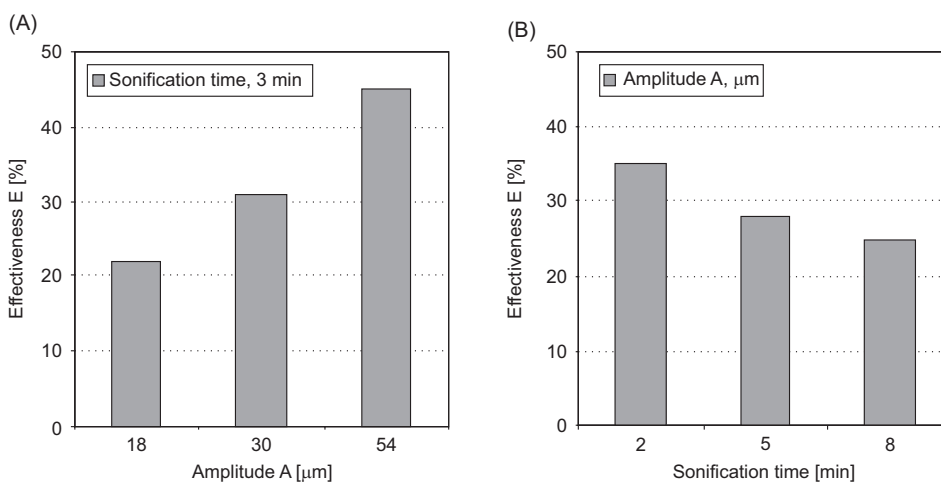


Fig. 1. The effectiveness of the TOC decreasing at the changeable vibration amplitude ($t = 3 \text{ min}$) (A) and at the changeable sonification time ($A = 18 \mu\text{m}$) (B)

increased their share in TOC even further. The effectiveness of sonochemical organic matter removal is confirmed also by the decrease in oxygen consumption. This indicator decreased with increasing vibration amplitude in the range examined. The most favourable effect was noted for the sample sonicated at the highest field intensity (for $A = 54 \mu\text{m}$) – a reduction in oxygen consumption by more than three units. The pH values of the water tested (with $\text{pH} = 7.92$) slightly decreased upon sonification and remained at a comparable level for the amplitude values examined. However, no effect of decreasing water contaminant concentration was confirmed in the UV_{254} absorbance determination results. The lack of relationship between the TOC determinations and the UV_{254} absorbance values found by other researchers, as indicated in the literature review, suggests different relationships to exist in the results of sonification of HA-prepared and natural water samples. In the second combination of parameters, fixed vibration amplitude of $18 \mu\text{m}$ and a variable sonification time were applied. Lengthening the sonification time decreased the effectiveness of the organic compound removal process, as measured by the TOC index. The best effectiveness equal to 35 % (a reduction in TOC by 5.13 mgC/dm^3) was obtained for the shortest sonification time of 2 minutes (Fig. 1B). The reduction of the DOC index (with a lower efficiency compared with TOC) increased again the share of dissolved carbon in the determination of TOC. Similarly as in the first parameter combination, a reduction in the oxygen consumption of sonicated water samples was noted with lengthening time. The decrease of oxygen demand in this experiment might indicate that the organic contamination was reduced by means of sonochemical oxidation. Increasing the sonification time, similarly as the amplitude, slightly decreased the pH of water. Favourable results of TOC reduction in natural water tested were obtained after a sonification time shorter than stated in the results for HA-prepared water samples, as presented in the literature review.

Further tests were aimed at the determination of the effect of the reaction of water sonicated ($\text{pH} \sim 3$ and $\text{pH} \sim 9$) on the variation of organic contaminant contents, as measured both directly as TOC and indirectly as oxygen consumption and UV absorbance. The results of tests for water samples with acid reaction (Table 2) do not confirm the favourable effect found at the natural water pH. Upon the application of ultrasounds the TOC index decreased only slightly. Its values remained, however, at

Table 2

Effect of amplitude and sonification time on the selected indicators of water (acid reaction)

Index	Before UD acid reaction	After UD					
		3 min			18 μm		
		18 μm	36 μm	54 μm	2 min	5 min	8 min
pH	3.01	2.99	2.97	2.96	3.0	2.99	2.98
TOC [mgC/dm^3]	10.27	9.68	9.80	10.0	10.22	9.87	9.90
DOC [mgC/dm^3]	6.94	—	—	6.55	—	—	6.69
Oxygen consumption [mgO_2/dm^3]	8.98	8.57	8.16	7.75	8.57	8.16	8.16
Absorbance UV_{254}^5	0.82	0.94	0.95	0.96	0.94	0.94	0.95

a comparable level, without any distinct relationships with time and amplitude. Water samples with a corrected pH value of $\text{pH} \sim 3$ were characterized by a smaller share of dissolved organic carbon in TOC concentration. This is associated with the solubility of humus substances, which increases with increasing water basicity.

In the subsequent phase of the experiment, the effect of basic water reaction on the effectiveness of sonochemical water HS removal was examined. For this purpose, surface water was alkalized to $\text{pH} \sim 9$. Similarly as for the natural and acidified water samples, the pH of the water sample was slightly decreased by ultrasonic processes. However, no favourable effects of organic water contamination removal was noted under these conditions, as indicated by the values of the TOC and DOC indices and UV_{254} absorbance (Table 3). At the same time, a reduction in water oxygen consumption was confirmed in the basic medium, irrespective of the ultrasonic parameter combination used. This might suggest an oxidation of inorganic compounds that are also covered by this indicator.

Table 3

Effect of amplitude and sonification time on the selected indicators of water (alkaline reaction)

Index	Before UD alkaline reaction	After UD					
		3 min			18 μm		
		18 μm	36 μm	54 μm	2 min	5 min	8 min
pH	9.04	9.00	8.98	8.91	9.00	8.98	8.91
TOC [mgC/dm^3]	11.73	14.54	14.66	14.23	13.59	14.01	14.10
DOC [mgC/dm^3]	8.08	—	—	9.40	—	—	10.36
Oxygen consumption [mgO_2/dm^3]	12.24	11.83	11.42	11.02	11.42	10.61	10.2
Absorbance UV_{254}^5	0.94	1.03	1.06	1.11	1.05	1.05	1.07

The analysis of the results obtained at different water pH values may lead to the conclusion that the form of occurrence of compounds prevailing in the composition of humus substances is crucial to the effectiveness of the process under examination. Water reaction influences also the intensity of radical reactions that provide a basic mechanism of sonochemical oxidation of organic contaminants. In the surface water examined, the process effects are largely associated with the susceptibility of fulvic acids and its compound to ultrasounds. The solubility of HS increases with increasing pH, which has an effect on the sonochemical oxidation results. At acid and neutral reactions, HAs can occur mainly in the form of colloids. It is only in a basic medium that they pass into dissolved forms. However, FAs, being prevalent in surface water, undergo dissociation already at a lower pH. Therefore, no decrease in organic contamination indices was noted at acid reaction, whereas favourable effects were observed at a slightly alkaline reaction of natural water of $\text{pH} = 7.92$. The increase of the amplitude (intensity) intensifies these effects in the water examined (Fig. 2A). A significant TOC reduction took place already within the first 2 minutes of sonification at an amplitude of 18 μm (Fig. 2B). On the other hand, no organic contaminant removal by the ultrasonic method occurred in a strongly basic medium ($\text{pH} \sim 9$). The basicity of water inhibits the processes of oxidation of the radical type. The acceptors of OH^\bullet

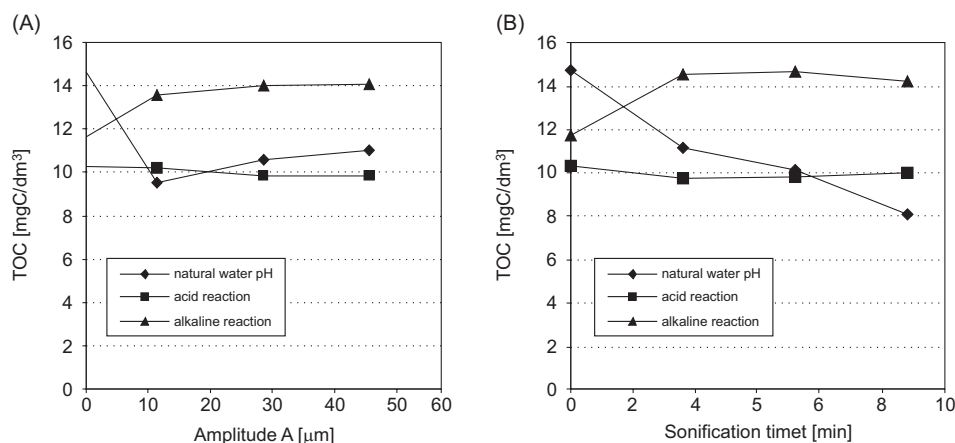


Fig. 2. The influence of the vibration amplitude ($t = 3$ min) (A) and the sonification time ($A = 18$ μm) (B) on the TOC index changes depending on the water pH

radicals are then mainly bicarbonate and carbonate ions (that determine the water basicity).

Summary

Organic contaminants, as measured by the TOC index, were removed from natural water to a different extent, depending on the ultrasonic field parameters and pH values applied. The effect of ultrasonic field intensity, as defined by the vibration amplitude, on process effectiveness was confirmed for water investigated. The maximum effectiveness of the ultrasonic method, *ie* 45 %, was achieved for the highest amplitude of 54 μm and short exposure time of 3 minutes. The effect of removing water contaminants in the sonochemical oxidation processes was indicated by a reduction in the oxygen consumption index. The effect of water pH on the process effectiveness remains disputable. From the preliminary investigation results provided herein it can be inferred that the medium of natural water pH is most advantageous. However, this does not resolve the question of validity of the obtained effect to other natural waters, *ie* those with a different pH. For establishing the pH range advantageous from the point of view of ultrasonic field application, investigation will be continued for surface waters with a different pH. To determine preliminary relationships, water samples prepared with fulvic acids, which are prevalent in the surface water medium, will be used. Further experiments will be aimed also at the determination of the effectiveness of the H₂O₂ and US hybrid process in the removal of organic water contamination.

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METODY SONOCHEMICZNE USUWANIA ZANIECZYSZCZEŃ ORGANICZNYCH

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Abstrakt: Wody powierzchniowe charakteryzują się zmiennością składu i stężeń organicznych zanieczyszczeń zarówno naturalnych, jak i antropogennych. Zanieczyszczenia i domieszki organiczne, na skutek przemian chemicznych wywołanych utlenianiem i dezynfekcją, mogą stać się prekursorami szkodliwych ubocznych produktów tych procesów (UPU/UPD). Usuwanie zanieczyszczeń organicznych jest więc istotnym problemem technologii wody, który dominuje w tematyce badawczej. Zmniejszenie organicznego zanieczyszczenia wody uzyskuje się metodą: koagulacji, adsorpcji, utleniania i procesów membranowych. Przewadzone są także badania nad możliwością zastosowania w tym celu także innych rozwiązań. Są one skierowane na tzw. metody hybrydowe, łączące metody chemiczne z czynnikami fizycznymi. Przykładem mogą być procesy pogłębnego utleniania, w których jako inicjatory rodników hydroksylowych wykorzystuje się promieniowanie ultrafioletowe lub ultradźwięki.

Badania literaturowe, stanowiące pierwszej część artykułu, wskazują, że niekonwencjonalna metoda ultradźwiękowa jest opisana na podstawie efektów uzyskanych dla wód preparowanych (najczęściej komercyjnymi preparatami kwasów humusowych). Z punktu widzenia składu substancji humusowych w wodach powierzchniowych (gdzie dominują kwasy fulwowe), uzasadniona jest weryfikacja tych efektów w środowisku wód naturalnych. W drugiej części artykułu przedstawiono wyniki badań dotyczące efektów stosowania pola ultradźwiękowego o dużym natężeniu i stałej częstotliwości 20 kHz. Substratem badań była woda powierzchniowa rzeczna pobrana z dwóch źródeł. Testy laboratoryjne prowadzono przy zmiennym czasie nadźwiękowania i różnej amplitudzie drgań. Kontrolę zmian zanieczyszczenia organicznego wody zapewniały głównie analizy wskaźnika OWO. Na początku eksperymentu i po zastosowaniu badanych metod sonochemicznych, wykonywano oznaczenia wybranych parametrów: OWO (TOC), RWO (DOC), utlenialności, UV_{254} i pH. Na ich podstawie oceniano efekty procesu w zależności od zmiennych parametrów ultradźwiękowych.

Słowa kluczowe: uzdatnianie wody, zanieczyszczenia organiczne, ultradźwięki