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ZOFIA IGNASIAK*

TECHNOLOGICAL ASSESSMENT OF THE WATER OZONATION PROCESS

The ozone decomposition in water has been discussed, emphasis being given to the role of hydroxyl radical in ozonation of water pollutants. It has been shown that the ozonation process may be optimized by choosing the condition that would prefer one of two mechanisms of oxidation reaction. The role of ozone as a technological factor in removal of many trace pollutants present in water and giving it advantageous organoleptic features has been discussed. The effects of water disinfection with chlorine and ozone have been compared, stating that the latter shows also viriocidal activity. In conclusions the criterion of the application of ozonation process in water treatment has been presented, attention being paid to the economic aspects of this process. The paper is based on the recent literature data and the results of investigations on water ozonation performed by the authoress.

1. MECHANISM OF OZONATION REACTION

Ozonation of water has become more and more widely used unit process of water treatment. According to the most recent information the world number of water treatment plants in which water ozonation is used exceeds one thousand. The chemism of ozone is not, however, recognized satisfactorily. Both the mechanism and kinetics of ozone dissociation in water have been studied for many years. It has been stated that due to the reaction catalyzed by hydroxide ions ozone is decomposed in water into radicals. Hydroxyl radicals produced during ozone degradation are particularly active oxydants. Very often they decide upon the oxidation of more stable substances present in water.

Hoigne and Bader [23, 24] have stated that ozonation of water may follow in two different courses. In low pH range dissolved substances, chemical groups with double bonds, organic chromophores and amine compounds react directly with ozone molecules. When the reaction conditions are chosen so that degradation of ozone precedes its reaction with organic compounds, hydroxyl radical becomes the most efficient oxidant.

* Research Institute on Environmental Development, Poznań Division, 60-613 Poznań, ul. Drzymały 24, Poland.

Although hydroxyl radicals are much less selective than the ozone molecules, their selectivity plays an important role in the course of oxidation process. They react particularly fast with aromatic hydrocarbons, alcohols, formic acid, and much more slowly with acetic and oxalic acids. Hydroxyl radicals may be trapped in reactions with carbonate and bicarbonate ions, which yield radicals HCO_3^- of low reactivity:

$$OH^{\cdot} + CO_3^{-} \rightarrow OH^{-} + CO_3^{\cdot},$$

$$OH^{\cdot} + HCO_3^{-} \rightarrow OH^{-} + HCO_3^{\cdot}.$$

Since the concentration of carbonates in the water being treated is usually high, it should be expected that the trapping of hydroxyl radicals by carbonates decides upon micropollutants removal. The carbonates acting, moreover, as "radical traps" stabilize the ozone concentrations in water. The detailed investigations perfoemed by HOIGNE and BADER [24] have proved that desintegration of ozone in water may be reduced ten times or even to a higher degree by as low concentrations of bicarbonates as several milimols per 1 dm^3 of water. Optimization of water ozonation processes should be accomplished considering the following points:

1. High stability of ozone, i. e. its stabilization with bicarbonates at low pH values should be attained when selective oxidability of ozone is to be utilized, e. g. disinfection in potable water or colour removal of water containing humic compounds. The addition of carbonates to ozonized water used for swimming pools is also advisable.

2. When the dissolved compounds are resistant to oxidation, ozone should be transformed into more reactive hydroxyl radical form (to get it in practice one increases pH of water). To prevent the trapping of hydroxyl radicals before their reaction with organic substances, the concentration of carbonates in water should be kept at a suitably low level; more advantageous results can be expected when water is decarbonized before ozonation.

Therefore it may be stated that ozonation may be optimized by applying different methods, depending on whether the reaction with ozone is to be accomplished directly or via hydroxyl radical.

2. THE EFFECT OF OZONE ON SUBSTANCES PRESENT IN NATURAL WATERS

The fact that in ozonation process water is not only fully disinfected but that its quality is also unproved gains a more and more importance. Due to its oxidizing activity ozone removes the substances responsible for the unpleasant water odour. It also oxidizes troublesome organic and inorganic trace pollutants frequently toxic, especially if they are able to accumulate in living organisms. A number of such substances are transformed or decomposed due to high oxidizing abilities of ozone.

In ozonation of coloured water humic substances are split at the hydrogen bonds (GABOWICZ [16]), hence only some of them are decomposed to carbon dioxide, and the basic mass remains in water in form of coloured crenic and hypercrenic acids. The subsequent degradation of humic compounds to carbon dioxide is possible at high ozone doses. While establishing ozone consumption (GABOWICZ [16]) it has been stated that on the average 0.18 mg of ozone is required to remove 1 degree of colour. The effect of colour removal depends among others on the composition of humic compounds. Increasing concentrations of fulvic acids at the presence of humic acids reduce the efficiency of colour removal and the doses of ozone should be augmented (KOWAL [35]). The efficiency of ozonation increases with the pH value, and decreases with the increasing temperature. In the above process the contact time of water with ozone of 6–8 minutes is long enough to state a significant reduction in colour intensity. In the presence of phenols during water ozonation humic compounds are oxidized after the oxidation of phenols (GABOWICZ, WRONCZINSKIJ [14]). During the water ozonation process colour removal is accompanied by removal of odours. Ozonation of water in which odour and taste are of natural origin results usually in their complete removal. In all the cases of the earthy, mouldy, and pharmaceutical odours, water ozonation yields satisfactory results. The same refers also to tastes and odours resulting from preliminary water chlorination; they almost always dissapear after the subsequent ozonation (GOMELLA [19]). It should be also added that due to a greater amount of dissolved oxygen the ozonized water takes a pleasant fresh smell and its colour has a blue shadow, in contrast to the yellowish tinge resulting from chlorination.

Ozonation is rarely used in oxidation of iron and manganese compounds since there are many less expensive methods. Thus, it is used in cases when traditional iron or manganese removal methods give no satisfactory results or when the taste, odour or colour of water should be removed simultaneously. Usually iron and manganese appear together in water. If they are present in form of inorganic compounds, then during ozonation first bi- and trivalent iron and then the manganese compounds are oxidized to tetra- and heptavalent compounds. When iron and manganese occur as organic complex or colloidal particles, the ozone oxidation results in their precipitation with a simultaneous colour removal. In this case good effects are obtained by an intense preliminary ozonation and a subsequent sand filtration or microstraining of flocculated coloured compounds. The above method is successfully applied in France (GOMELLA [19]).

Cyanides occurring in water because of their toxicity are particularly threatening to the people's health. The efficiency of cyanides removal increases due to catalysts, especially Cu^{2+} (KOROLIEW, BOGDANOW [34]). While investigating the kinetics of their removal by means of ozone TYLOR et al. [50] have stated that oxidation of cyanides occurs in two stages. In first stage cyanides are oxidized to cyanates, and in the second stage the latters are oxidized to bicarbonates and nitrogen. The concentration of CN^- decreases proportionally to the ozonation time and ozone concentration. Rate of cyanides oxidation depends to a high extent on the pH value and is the highest at pH 10–11.

The investigations on hydrogen sulphide have shown that during ozonation it is oxidized to sulphates (JÜRS [31]; REISSANS, RUMMEL [44]); nitrites are converted into nitrates (JÜRS [31]; KEWZMANN [32]).

The behaviour of ammonium ion with respect to ozone is still a controversial problem. The investigations performed by REISSANS and RUMMEL [44] have not shown the degradation of ammonium by ozone. NOWAK [41] presented the formula for the oxidation of ammonium to nitrate by ozone. SINGER and ZILLI [46] have stated that the above process requires an elevated pH value; it is about 10–12 times more efficient at pH 9.0 than at pH 7. EICHELSÖRFER and HARPE [11] and GAD and COLUMBUS [17] have proved that reaction of ozone with ammonia in water solutions yields nitrates, whereas the ammonium ion is not oxidized since the pair of electrons at nitrogen is occupied.

Ozon — as one of the strongest oxidizing agents — decomposes a number of organic compounds among which the most susceptible to degradation are those having one or more nonsaturated bonds.

One of the most important activities performed by ozone is oxidation of phenols and chlorophenols. Many research works have been devoted to the degradation of those compounds by ozone, that is why the mechanism of ozonation with respect to phenols is relatively well known. The oxidation of phenol with ozone may result in the following products: muconic acid, glyoxal, oxalic acid, and carbon dioxide (GAD, COLUMBUS [17]; EISENHAUER [12]). Since the oxidation of phenols to carbon dioxide is accompanied by the inadequate increases in the ozone dose and its high losses, it is assumed (EISENHAUER [12]) that because of economic reasons this process should not be led to the very end. Kinetics of phenol oxidation with ozone is discussed in the paper by BAUCH and BURCHARD [1] who have stated that first benzene ring is disintegrated at the the dose of 3 moles of ozone per 1 mole of phenols. The subsequent formation of organic and inorganic acids requires 2.5 moles of oxygen (from ozone) for 1 mole of phenol. At the same time the authors have stated that the cresols react more readily with ozone than phenol. pH of water plays an important part in oxidation of phenols. At higher pHs the rate of phenol oxidation is also higher; the optimum pH ranges within 11.5–12.5 (KOROLIEW, BOGDANOW [34]).

The investigations on the application of ozone in removal of surfactants gave promising results. At suitably chosen parameters of the process (pH, contact time) high efficiency of removal has been obtained, especially in ozonation of higher concentration of surfactants. According to ZDYBIEWSKA and GROMADZKA [55] after 30 minute ozonation of sulphoxylate solution in concentration of 10 mg/dm^3 the oxidation efficiency amounted to 90.2%, and in case of sulphonic salt of alphenol it amounted to 72.3%. For 4 different anion active surfactants being oxidized under the same conditions GROSSMAN [20] has stated the removal of those substances ranging from 78.5 to 98.1%. In the recently accomplished investigations performed by the Poznań Division of the Research Institute on Environmental Development (IGNASIAK and coworkers) in a dynamic flow system on pilot station TRAILIGAZ it has been stated that within low concentrations of surfactants (0.5–2.0 mg/dm^3) the effects of oxidation with ozone amounted to 80–90% for cosulphonate and 80–84% for deterlone. The best oxidation effects have been obtained for neutral pH of water at 10 minute contact time (figs. 1, 2). French investigations (MIGNOT [39]) give information that in ozonation of surfactants there exists a limit value to which the amounts of these substances can be reduced, which is equal to 60–90% of the initial value, and that these results cannot be improved even by increasing the dose of ozone.

Tests (performed on white mice and rats) (KOROLIEW, BOGDANOW [34]) on the degradation products of surfactants formed during water ozonation have proved that they

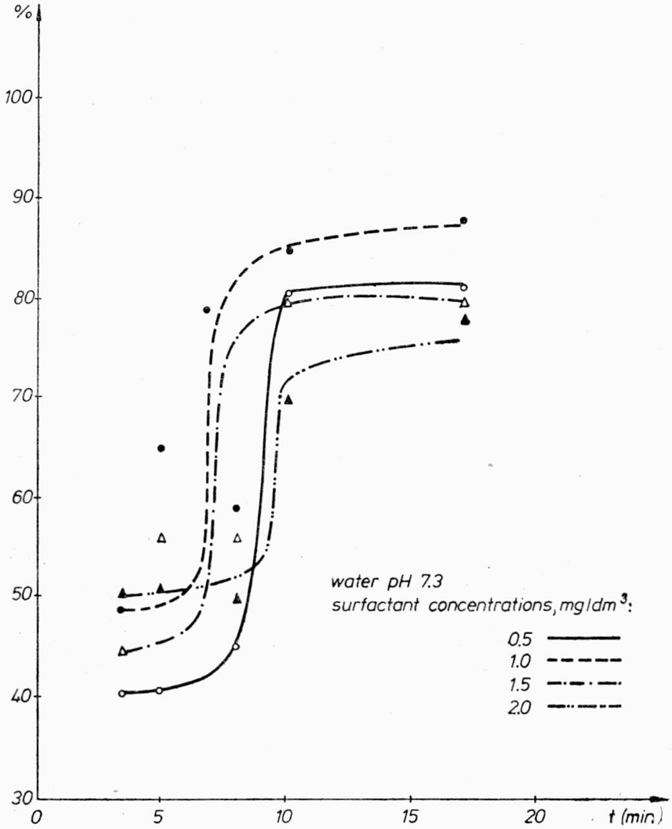

Fig. 1. Cosulphonate ozonation at different ozone doses

Rys. 1. Utlenianie kosulfonatu przy różnych dawkach ozonu

are less toxic and show weaker tendency for accumulation in organisms than the initial substrates. It has been also stated that the products of ozonation have no allergogenic properties.

During ozonation of water one also observes the removal of PAH which are classified among carcinogenic compounds. PAH being not readily soluble in water are found in higher concentrations in the presence of surfactants or are adsorbed on suspended solids. The resistance of PAH to ozonation is different. ILNITSKII et al. [30] have stated that the most resistant is benzo(a)pyrene (B(a)P) considered to be the strongest carcinogenic agent. It has been also stated (REIHERT [43]) that the degradation of B(a)P is strongly influenced by micropollutants present in water. During the investigations carried out in the Poznań Division of the Research Institute on Environmental Development (IGNASIAK and coworkers, 1977) B(a)P in concentration of 30–300 ng/dm^3 was removed in 82–93%. Fluorantene in concentration of 80–1200 ng/dm^3 was removed in 67–87%. The oxi-

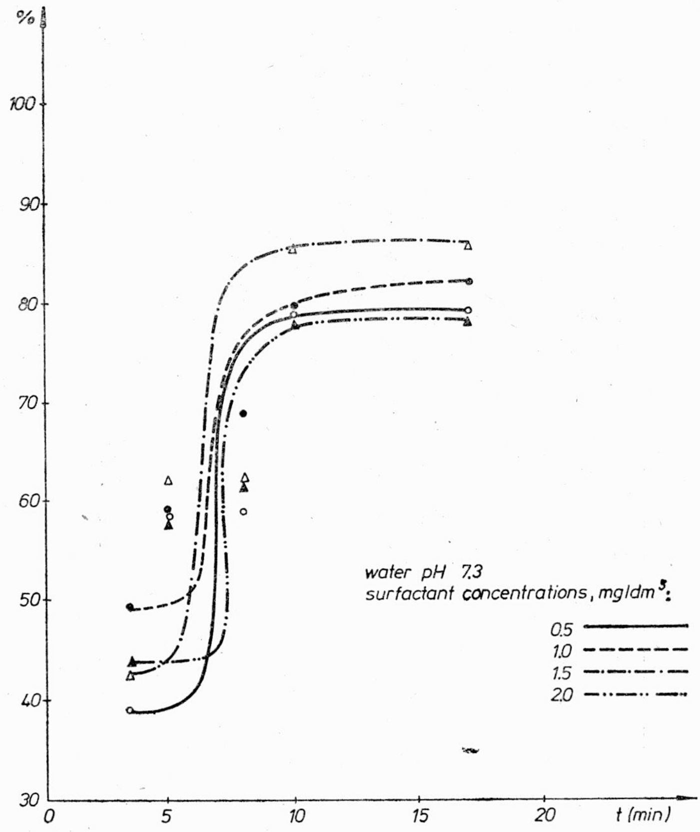

Fig. 2. Deterlone ozonation at different ozone doses

Rys. 2. Utlenianie deterlonu przy różnych dawkach ozonu

dation effects depend on the ozone dose, contact time, and pH of water. For PAH removal the recommended contact time should be equal to 5 minutes and pH neutral (figs. 3, 4). It has been also stated that the presence of surfactants in water increases the effect of PAH oxidation by 17%. So far the products of PAH oxidation has not been known. Since they are suspected to be strongly toxic, water after ozonation should be subject to adsorption on activated carbon.

The action of ozone on pesticides is different, depending on their composition and structure. Chloroorganic pesticides (lindan, dieldrin, DDT, and γ-HCH) are hardly attacked by ozone (MIGNOT [39]). As far, however, as the oxidation of phosphoorganic pesticides is concerned (e. g. parathion and malathion) a strong oxidizing activity of ozone has

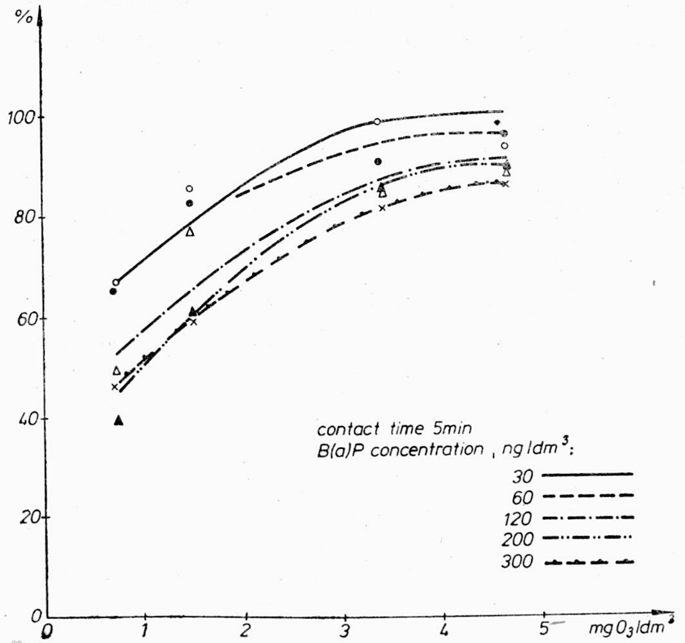

Fig. 3. B(a) P ozonation at different ozone doses

Rys. 3. Utlenianie B(a)P przy różnych dawkach ozonu

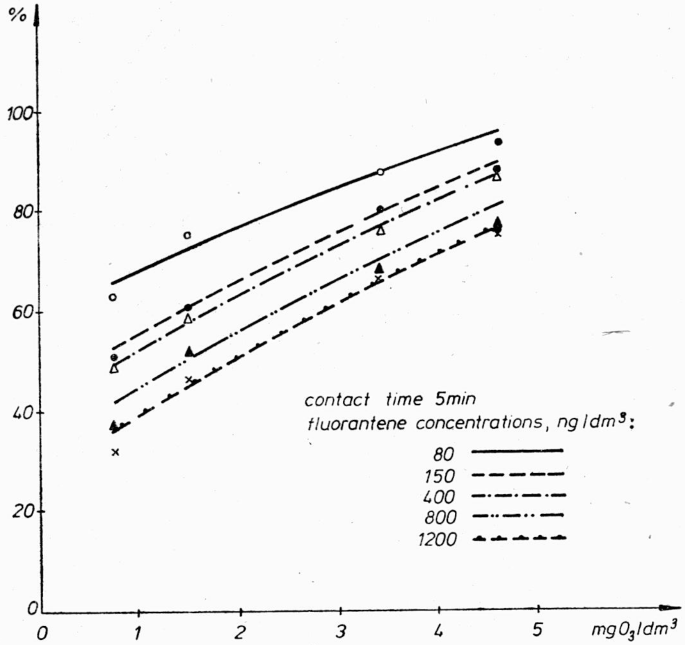

Fig. 4. Fluorantene ozonation at different ozone doses

Rys. 4. Utlenianie fluorantenu przy różnych dawkach ozonu been stated (KOROLIEW, BOGDANOW [34]; GABOWICZ, WRONCZINSKIJ [14]). The ozone doses required for oxidation of phosphoroorganic pesticieds exceed 45–75 times their concentrations in water (MIGNOT [39]) (fig. 5).

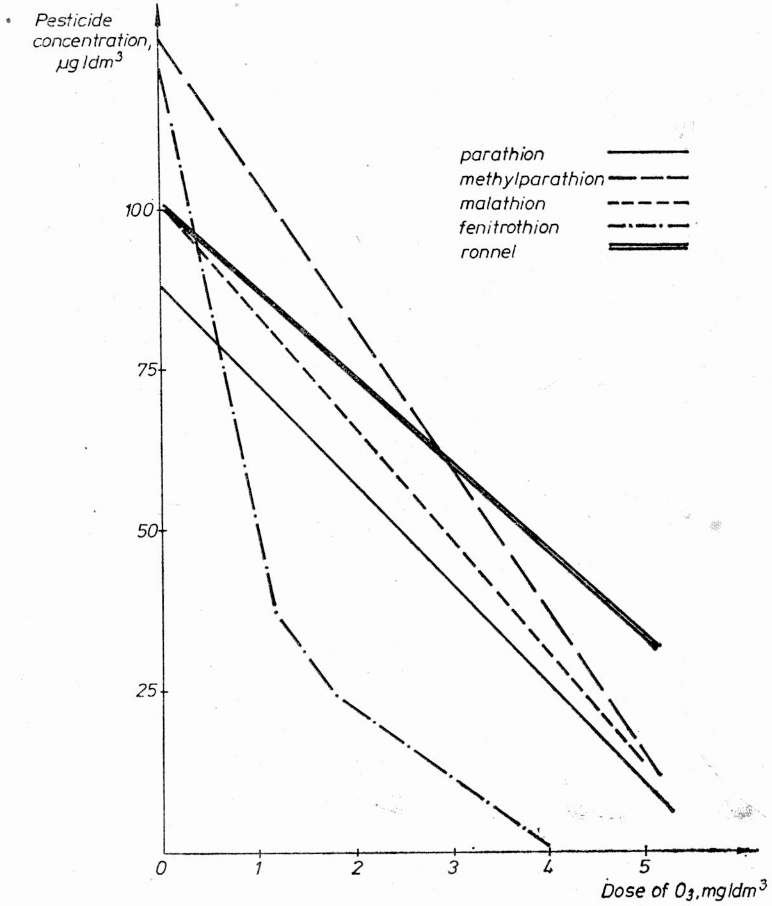

Fig. 5. Ozonation effect on the reduction of pesticide concentration according to DEGREMONT [39]
Rys. 5. Wpływ ozonowania wody na obniżenie zawartości pestycydów według DEGREMONTA [39]

3. THE EFFECT OF OZONATION ON COAGULATION PROCESS

In sixties of our century, la Compagnie des Eaux et de l'Ozone introduced and patented a new M-D method (micellarization and demicellarization) of ozone application as a coagulation aid (GUILLARD [22]). This method is based on a specific action of ozone on colloidal particles yielding flocculation of organic macromolecules responsible for colour and turbidity of water. The method raised a great interest, as it allows to eliminate both the sedimentation tank and the equipment for dissolving and mixing coagulants with water. In the M-D method ozonation is most frequently combined with microfiltration. This method found its application in a few water-treatment plants as it requires a low turbidity of water and high ozone doses. Promising effects, however, may be obtained by combining the M-D method with conventional coagulation (LATAWIEC [37]).

According to SONTHEIMER [47] the application of ozone as a flocculant is advisable only when viriocidal effects and improvement of water taste and odour are to be obtained.

4. EVALUATION OF WATER OZONATION AND CHLORINATION FOR THE PURPOSE OF DISINFECTION

Bacteriocidal properties of ozone have been known for a long time. The studies on mechanism of ozonation of bacteria have shown that ozone reveals its destructive activity at suitably long contact time and high enough concentration in water. This activity consists in catalytic oxidation of protein mass (GOMELLA [19]). According to YAO [53] the disinfecting efficiency of ozone referred to that of chlorine amounts to 2.65.

In water bacteriocidal action of chlorine occurs in two stages: 1. diffusion of chlorine through the cell wall, 2. reaction with intercellular enzymes resulting in the inhibition of cell metabolism (SPANDOWSKA [48]). The full bacteriocidal effect is obtained with the dose of chlorine that ensures the concentration of 0.5 mg of chlorine/dm^3 after a half an hour contact.

Quite recently it has been stated that ozone is a particularly effective with respect to viruses, to which chlorine was almost powerless. This is probably due to a characteristic tendency of viruses to form agglomerationsof different sizes through which chlorine cannot penetrate. Hence the survival of viruses is observed even at high concentrations of chlorine in water.

The studies on viriocidal effects of ozone performed by COIN et al. [7] at the Institute of Pasteur have shown that the virus of poliomyelitis is completely inactivated during 3–4 minutes at the ozone dose of 0.4 mg/dm^3. A full viriocidal effect is guaranteed by the ozone concentration of 0.4 mg/dm^3 remaining for the contact time of 8 minutes. By applying the above parameters one obtains at the same time the optimal removal of micropollutants. Contact time longer than 8 minutes appears to be ineffective and noneconomic (COIN [6]). Virostatic properties of ozone are similarly evaluated by American researchers (MCBRIDE [40]), according to whom contact time of 5 minutes is sufficient to neutralize completely bacteriophages f_2 used as model viruses.

This method of water ozonation is frequently criticized, as the ozone remaining in the water-distribution system quickly disappears and the water, even perfectly disinfected, may become liable to the secondary infection. Hence a combined method, ozonation and chlorination, is frequently applied in the technology of water treatment. In the light of the proceedings of conferences and opinions of some users (Hollopeau, Scheller [25]; Schalekamp [45]; Duszyński [10]) this problem may be presented as follows:

1. Well ozonized water does not show the possibility of a secondary infection in the water-distribution system. The respective investigations performed in St. Gallen and Częstochowa have shown that any difficulties connected with the secondary infection of water appeared when it was not sufficiently treated prior to ozonation or the ozone dose was too low.

2. The state of water-distribution system plays an important part: the experiments carried out in Nice and Paris (Girardot [18]) have shown that the bacterial growth is not related to the absence of ozone in the system. Moreover, Hollopeau and Scheller [25] have shown that the presence of residual ozone either does not guarantee the absence of spores and that the coliforms are present also in residual chlorine containing waters since a small amount of free chlorine cannot prevent the secondary infection of water. Probability of spreading virus diseases through the municipal water-distribution system was several times questioned (Coin [6, 7]). Nevertheless, some epidemies of viral diseases (viral hepatitis, poliomyelitis) were accompanied by presence of the above-mentioned viruses in drinking water.

Insufficiently treated municipal water taken from the polluted surface water resources is potentially dangerous. It has been observed that the infections with poliomyelitis virus show an increasing tendency in summer and autumn months in the regions supplied with surface waters, the phenomenon being unknown in case of ground water intakes (Coin [8]). Mass epidemies of viral hepatitis reported by Clark and Chang [5] and Taylor [49] are due to the same reasons.

The parameters determining distinctly the purity of water with respect to viruses are: turbidity, total bacterial count, and coliform number. In the clear water the number of bacteria is usually small, and the cases of infection with viral hapatitis are seldom reported (Spandowska [48]).

5. CRITERIA OF THE APPLICABILITY OF WATER OZONATION

The existing opinion about the usefulness of ozone for water works as well as the described universal advantages of water ozonation may lead to conclusion that in all the water treatment plants special equipments for water ozonation should be installed. The decision, however, must be based on rational technological and economical premises, since ozonation is not always necessary.

There is no model scheme for the application of ozonation method. Its advisability should be thus considered in cases when conventional methods of water treatment and disinfection do not assure the required quality of drinking water. It should be born in mind that ozonation of water is expensive because of high capital and exploitation costs, chiefly resulting from the uptake of electric energy.

In view of the literature review presented in former sections as well as basing on the experiments performed in the Poznań Division of the Research Institute on the Environmental Development (SPANDOWSKA [48]; IGNASIAK, BIŁOZOR [28]; MASTALERZ et al. [38]; BŁAŻEJEWSKI et al. [4]; IGNASIAK et al. [29]), it may be stated that the decision about water ozonation must be based on the following criteria:

1. the necessity of application of ozone as viriocidal agent,
2. its application as water treatment aid.

The first criterion of ozone application refers to surface water intakes polluted with municipal wastewaters. Since ground and infiltration waters do not contain viruses, their penetration into ground being possible to the distance not greater than 5 meters.

When surface waters are taken for potable purposes, the whole process of water treatment should be strictly controlled, especially at high raw water turbidity, suspended solids and organics concentrations. Viruses coated with a layer of organic pollutants are to a high degree protected against the action of even high concentrations of ozone.

Application of ozone as a technological factor should be considered in cases when the raw water in certain periods has unpleasant taste and odour, caused by plankton growth, which cannot be removed with chlorine dioxide, potassium permanganate or sorption on activated carbon. Ozonation is, moreover, necessary in case when chlorination of water impairs its taste and odour. It is also necessary at the presence of phenols, surfactants, pesticides, and PAH recognized as the carcinogenic compounds, if their concentrations exceed the permissible standards (IGNASIAK, BIŁOZOR [28]). When applying ozonation it should be remembered that good effects are obtained when the water is adequately pretreated. This, in particular, refers to the turbidity removal. Considering the products resulting from water ozonation wich are not yet identified the process should be combined with the activated carbon adsorption.

Decisions about the treatment phase in which ozone is to be introduced, its dose, its possible splitting into two-stage dosage, eventually a combined application of ozone and activated carbon adsorption should be made individually for each water in order to choose the most advantageous system.

6. SUMMARY

The paper is based on literature review and on the analysis of the studies of water ozonation performed by the authoress. The following problems have been discussed: the mechanisms of water ozonation and the influence of ozone on the natural water pollutants and on the coagulation of water. The processes of water ozonation and chlorination have been, moreover, evaluated and the criteria of water ozonation discussed.

The role of hydroxyl radical was emphasized showing that the ozonation process may be optimized by choosing the conditions preferring one of two mechanisms of ozone reaction with water components.

The effect of ozone on the particular trace pollutants occurring in water has been discussed, and the role of ozone as a technological factor in the water treatment underlined. While estimating the ozonation and chlorination of water for disinfective purposes, the priority of ozonation has been shown due, among others, to its viriocidal activity.

It has been stated that the introduction of ozonation into technology of water treatment is justified by its viriocidal activity or by its action as a technological aid in the removal of micropollutants. Since chlorine is an adequate disinfective agent in case of bacterial pollution only when chlorination of water deteriorates its taste and odour or results in formation of toxic compounds (e. g. halogenomethanes) application of ozonation is fully justified.

Attention was paid to a strict control of water treatment process, since only well pretreated water quarantees good effects of ozonation. Having in mind ozonation products not yet identified it has been stated that the process should be considered in combination with activated carbon adsorption. It has been emphasized that ozonation requires high capital and operation expenditures chiefly due to the electric energy intake. That is why the decision about application has to be fully justified.

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WPŁYW OZONOWANIA NA WYBRANE SKŁADNIKI WÓD I PROCESY JEJ UZDATNIANIA

W artykule omówiono reakcje rozkładu ozonu w środowisku wodnym podkreślając rolę rodnika hydroksylowego w procesach utleniania ozonem domieszek wody. Wskazano na sposób optymalizacji procesu ozonowania przez dobranie warunków preferujących jeden z dwóch mechanizmów reakcji utleniania. Omówiono działanie ozonu jako czynnika technologicznego w usuwaniu wielu zanieczyszczeń śladowych występujących w wodzie i nadawaniu jej korzystnych cech organoleptycznych. Dokonano oceny porównawczej dezynfekcji wody chlorem i ozonem wskazując na wyższość ozonu polegającą m.in. na jego wirusobójczym działaniu. We wnioskach przedstawiono kryteria stosowania procesu ozonowania w uzdatnianiu wody i zwrócono uwagę na ekonomiczne aspekty tego procesu. Artykuł jest napisany na podstawie piśmiennictwa z ostatnich lat i analizy wyników własnych badań z ozonowaniem wody.

DER EINFLUSS DER OZONISATION AUF AUSGEWÄHLTE WASSERINHALTSSTOFFE UND AUFBEREITUNGSVERFAHREN

Besprochen werden die Zerlegungsreaktionen des Ozons im Wasser, wobei die Rolle des Hydroxyls in der Oxydation von Wasserinhaltsstoffen besonders hervorgehoben wird. Aufgezeigt wird eine Optimierung der Ozonisation die durch die Annahme eines der zwei bevorzugten Reaktionsmechanismen erreicht wird. Als technologischer Faktor wirkt Ozon auf den Abbau zahlreicher Spurenverunreinigungen die im Wasser vorkommen ein und verbessert wesentlich den Geschmack des Wassers. Ein Vergleich der Desinfektion des Wassers mit Hilfe von Chlor und Ozon, ergibt bessere Ergebnisse bei der Ozonisation die u. a. auch Viren desaktiviert. In den Folgerungen werden die Anwendungskriterien der Ozonisation zur Wasseraufbereitung erörtert, wobei ökonomische Aspekte auch in Betracht gezogen wurden. Dem Beitrag lagen neuere Ergebnisse aus dem technischen Schrifttum letzter Jahre sowie eigene Untersuchungen des Verfassers zugrunde.

ВЛИЯНИЕ ОЗОНИРОВАНИЯ НА ИЗБРАННЫЕ КОМПОНЕНТЫ ВОД И ПРОЦЕССЫ ЕЁ ПОДГОТОВКИ

В статье обсуждены реакции разложения озона в водной среде, подчёркивая роль гидроксильного радикала в процессах окисления озоном примесей воды. Указывается способ оптимизации процесса озонирования посредством подбора условий, отдающих предпочтение одному из механизмов реакции окисления. Обсуждено действие озона как технологического агента в удалении многих следов примесей, выступающих в воде, а также придание ей полезных органолептических свойств. Произведена сравнительная оценка дезинфекции воды хлором и озоном, указывая на преимущество озона, заключающееся, наряду с другими, в его действии, уничтожающем вирусы. В выводах приведены критерии применения процесса озонирования в водоподготовке и уделено внимание экономическим аспектам этого процесса. Статья написана на основе литературы последних годов и анализа результатов собственных исследований по озонированию воды.