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WATER TREATMENT USING CHLORINE DIOXIDE WITH CONTROLLED DOSING AND SIMULTANEOUS CONCENTRATION MONITORING

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ABSTRACT
This article describes the real model of water treatment using the chlorine dioxide generator with online continuous monitoring of the real concentration of ClO_2 in the drinking water line to keep the right chlorine dioxide concentration for effective disinfection effect. There are two issues pointed out, first the disinfection effect of clorine dioxide and as second the effect on the biofilm that is the ideal platform for the bacteria growing. A part of arrticle is the expertise of ClO_2 effect on biofilm, results from testing of the analyser which is set to monitor the concentration of chlorine dioxide and practical experiences with the mentioned disinfection system.

KEY WORDS
Chlorine dioxide, Biofilm, Disinfection, Water treatment

Introduction

Water as the most important factor of human life and whole nature is with growing of negative affect of industry and consum way of our life absorbs all the harmfull substances and energies. Therefore, there is taken a considerable effort to find out new and more efficient technologies to keep the water in its original condition ,chemical or structural, to develop new ways of thinking to invent as simple and as efficient technologies, as possible. Nowadays, the science is moving on the way of structural conditioning of the water, what is one of the very important factor, which can give the water different properties regarding to effect to human health and condition. Regarding to biological screen of water, as one of the most important its quality factor, there are several disinfection methods, more or less efficient, but in practise, in relation to side effects, the disinfection with chlorine dioxide is the most efficient one. Not only because of its fast and efficient disinfection effect but also the anti-biofilm effect as well. Chlorine dioxide is very effective against very dangerous kind of bacteria called Legionella. Moreover, using this kind of water treatment technology, we can safe cost for energy need for heating of the water to get the proper temperature for reliable disinfection effect on all points of the water lining.Chlorine dioxide is of course a very toxic and dangerous substance, therefore, it is being produced in the ClO_2 generators and applied, using special dosing system, continuously in time in small amounts directly to the system. The procedure to be monitored and dosing to be controlled according to the real concentration of ClO_2 in the water line. Concentration of ClO_2 in the line must be kept in proper range to keep reliable disinfection effect and avoid harmfull effect on the lining material or human health. Anyhow, there is a pressure fall in several bars at the tap point of the lining and the diluted chlorine dioxid sublimies to atmosphere.

Drinking water safety

Safe drinking water, as defined by the Guideliness, does not represent any significant risk to health ocer a lifetime of consumption, including different sensitivities that may occur between life stages.[1]Risk characterization includes understanding of data completeness, estimating the “potential” distribution and abundance of invasive species, estimating the potential rate of spread of species, and estimating their probable risks, impacts, and costs.[2]Legionella pneumophila , the agent responsible for Legionnaires’ disease, has been respon- sible for the deaths of countless number of individuals worldwide since its association with human illness was first established in 1976.The omnipresence of Legionella within water systems presents a potential health concern, particularly for immunocompromised persons, in both developing and industrialised nations. The primary aim of this study was, therefore, to present a simplistic quantitative microbial risk assess- ment (QMRA) for Legionella within water distribution systems.[3]Legionnaires disease is an atypical pneumonia

[1]WHO, Geneva 2004.Guidelines for Drinking-water Quality, 3rd edition. ISBN 92 4 154638 7. p.1.
[2] Stohlgren - T.J., Schnase , J.L.., 2006: Risk Analysis, Vol. 26, No. 1, 2006. Risk Analysis for Biological Hazards: What We Need to Know about Invasive Species.p. 167.
[3] Storey, M.V. - Ashbolt, N.J. - Stenström, T.A., 2004: Biofilms, thermophilic amoebae and Legionella pneumophila – a quantitative risk assessment for distributed water.

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caused by bacteria of the genus *Legionella.*[4] On account of its tolerance to heat and low nutritional needs, *Legionella pneumophila* is widespread in many water systems and therefore poses a serious public health risk. The micro-organism passes from its natural reservoirs into the water distribution network, where the main source of contamination can be found. Pneumonia from *Legionella* is caught by inhaling particles of aerosol emitted by showers, taps, cooling towers, as well as by hydromassage tubs, spa waters, ornamental fountains etc. In hospitals micro-aspiration may be an important route of transmission for patients lacking normal respiratory reflexes.[5]

Very important factor regarding to bacteria grow are biofilms. Biofilms are the product of adhesion and growth of micro-organisms on surfaces. On the one hand, biofilms act as biological filters by mineralizing biologically degradable material from the water and forming locally immobilized biomass. On the other hand, biofilms may unpredictably emerge in the distribution system and may cause diverse problems in terms of bacterial contamination with hygienically relevant bacteria or spontaneous increases of bacterial cell counts in bulk water. Thus, they represent an important factor for the stability of drinking waters. The drinking water distribution net offers a very large surface area for the adhesion of bacteria. At least 95% of the total bacterial biomass in drinking water are found on the walls of the distribution system.[6]

Water threatment metods

The use of water treatment technology is not new, but dating back 6000 years when the Greeks used charcoal filters, boiling, straining and exposure to sunlight to improve the aesthetic quality of drinking water.[7] Although disinfection methods currently used in drinking water treatment can effectively control microbial pathogens, research in the past few decades have revealed a dilemma between effective disinfection and formation of harmful disinfection byproducts (DBPs). Chemical disinfectants commonly used by the water industry such as free chlorine, chloramines and ozone can react with various constituents in natural water to form DBPs, many of which are carcinogens.[8]Chlorine is one of the most commonly used disinfectants for water disinfection. Chlorine can be applied for the deactivation of most microorganisms and it is relatively cheap.[9]Alternatives to chlorine as a primary disinfectant exist. Ozone is a very effective disinfectant, and where it is used for other purposes, usually for removal of organic micropollutants such as pesticides, it provides benefits in terms of reducing the microbiological challenge to downstream disinfection. However, ozone also forms by-products, particularly bromate. Chlorine dioxide is used as a primary disinfectant and in distribution worldwide, but there are limitations to its use because of the inorganic by-products chlorite and to a lesser extent chlorate. Where these chlorite by-products are elevated consequent to high ClO_2 doses, an additional chemical dosing process is required involving the addition of ferrous salts to reduce levels to below the WHO guideline limit of 0.7mg/l.[10]

Chlorine dioxide

ClO_2 exists as a stable free radical with an unpaired electron and reacts with organic and inorganic compounds mainly through a one-electron transfer reaction.[11]Chlorine dioxide, prepared early in the 19th century, was used in the treatment of water supplies in Europe after 1850. It was not until the 1940's, however, that experimental data on its bactericidal efficiency became available. Although chlorine and chlorine dioxide are similar in many respects, including the fact that both are powerful oxidizing agents, ClO_2 has 2.5 times the oxidation capacity of Cl_2. It was this feature that recommended ClO_2 for the control of odors and tastes in water supplies.[12]High efficiency of disinfection effect is shown on Figure 1.

[4]Fraser, D.W. – Tsai, T.R. – Orenstein, W. et al., 1977: Legionnaires' disease: description of an epidemic of pneumonia , N Engl J Med , 1977 , vol. 297. p. 1189 -97

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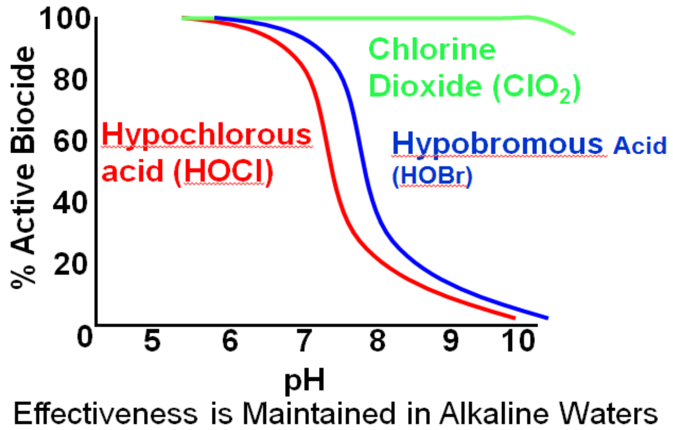

Figure 1 Disinfection efficacy of ClO2 according to pH[13]

Influence of temperature

Legionellae have been isolated from hot-water systems up to 66 ºC; however, at temperatures above 70ºC they are destroyed almost instantly (Dennis, Green & Jones, 1984; Dennis, 1988b). Kusnetsov et al. (1996) found that growth of all strains tested decreased at temperatures above 44–45 ºC, with the growth-limiting temperature being between 48.4 ºC and 50.0 ºC. The Legionella strains studied produced carbon dioxide up to 51.6 ºC, suggesting that some respiratory enzymes survive at this temperature. Complex water systems, such as warm-water plumbing systems, air-conditioners and hot tubs (also known as spa pools), are increasingly using water in the temperature range that encourages Legionella growth. In addition, these water systems can potentially produce aerosols, increasing the spread of the bacteria.[14]

Cost saving by using lower water temperature for warm water supply

The most common method for water disinfection is to heat up the warm water supply up to > 70 °C, to eliminate the bacteria's (special Legionellas). But a water temperature in range of 50 – 55 °C is enough in most applications. The common method needs a lot of energy, which is in general not necessary. The method is not secure and there is no reliability that on any tap the correct temperature is available. A high water temperature in water supply system is no guarantee for a 100 % disinfection. There is still a risk of disinfection with Legionellas.

The following example shows, how much energy is wasted and how much costs one can save by using lower temperature:

If you have i.e. a daily water consumption of approx. 50 m^3 and you have to increase your warm water system temperature in an amount of 15 °C from 55 °C up to 70 °C. In this case you receive by a gas price of 0,40 Euro/m^3 the following calculation:

- 40 % of total water consumption is warm water (in this case 20 m^3 /day).
- To increase the temperature of 1 m3 water in amount of 15 °C you need approx. 17,5 kW energy respectively 1,9 m^3 gas (calculated by heating value of gas).
- To heat 20 m^3 of warm water you need daily 38 m^3 gas.
- This is a high amount of energy you can save daily 15,20 Euro and 5548 Euro per year.[15]

Effect on biofilms of chlorine dioxide dosed at 0,2 mg/L by degerming units "Aquacon WH01" and "Aquacon CLO2"

Biofilms in the drinking water-distribution systems could release high concentrations of microorganisms to the streaming water. We tested the effect of chlorine dioxide dosed at 0,2 mg/L by the degerming units "Aquacon WHO1" and "Aquacon CLO2" on the biofilms and the levels of the released free floating microorganisms in water. The system used for the tests was the silicone tube model, which has been developed for this type of testing by the Institute of Hygiene and Public Health in Bonn. The test system consisted of silicone tubes, 4 mm in diameter, through which treated and/or untreated water flowed. The silicone tubes themselves were either fresh ones with no biofilm attached to the inner surface, or older ones which had been plugged in to the water system for several years and which contained well developed biofilms on their inner surface. The presence of microorganisms in the biofilms or of those released from the biofilms to the flowing water was tested for by standard microbiological cultivation methods. The presence and appearance of the biofilms was tested

[13] Advanced Water Technologies. - [on-line] available on - URL: https://www.prowater.co.uk/chlorine-dioxide-disinfection.html
[14] World Health Organization. 2007. Legionella and the prevention of legionellosis. ISBN 92 4 156297 8, p.30.
[15] Einsparpotenzial40415. *Internal documentoflotronicProcess Monitoring, Niederlassung Deutschland.*

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for by means of a scanning electron microscopy. The final concentration of chlorine dioxide of 0,2 mg/L was that prescribed by the “Drinking Water Regulation". The concentrations of microorganisms in the biofilms were determined in terms of Qolony Eorming Qnits (CFU) per unit area (CFU/cmz), while the concentrations of the microorganisms washed out from the biofilms to the flowing water were determined in terms of CFU/cm^2/sec or CFU/ml. The samples were collected once a week over the period of 70 days. The washout of bacteria from mature biofilms washed in untreated water displayed the average level of 1450 CFU/ml. After switching the system to the chlorine dioxide-treated water the level of washout dropped to zero. The presence of chlorine dioxide in the water also displayed the same effect on the levels of microorganisms in the biofilms, albeit over a longer time scale. After 70 days of continuous wash of the biofilms in the treated water the level of microorganisms in the biofilm dropped from the original 10.000.000 CFU/cm^2 to zero. In the tubes with the same biofilm but washed in untreated water the washout remained at 10.000.000 CFU/cm^2 over the whole test period. In the fresh silicone tubes with no biofilm at the beginning of the test period, the effect of washing in with the treated water was a complete suppression of the biofilm formation. When the same fresh silicone tube was washed in untreated water, a biofilm was formed with the level of microorganisms at 388.000 CFU/cm^2. All these findings were confirmed by the scanning electron microscopy.Supplementary suspension tests with bacterial colonies (Legionella pneumophila, Pseudomonas aeruginosa, Escherichia co/i and Enterobacter c/oacae) exposed to 0,2 mg/l of chlorine dioxide revealed that 5 minute contact time brought the level of bacteria under the detection level. Chlorine dioxide has been known for many years as a proven and very efficient disinfectant. However, till now it was not possible to apply this disinfectant in the fields of drinking water, warm water and Legionella-containing water disinfection because no methods for monitoring concentration of chlorine dioxide (0.05-0,2 mg/l prescribed by the “Drinking Water Regulation”) were available. The levels of chlorine dioxide in water can now be precisely monitored with the device “Aquacon CLO2". The selective measurements are based on a patented “lotronic” method. Tests performed at the Institute of Technical Chemistry at the Hannover University confirmed the suitability of the lotronic method for an automated monitoring of the chlorine dioxide levels in water. In comparison to the common laboratory methods (DIN EN 12671), the lotronic method, when applied for measurements of concentrations relevant in the drinking water treatment (0.1-0.3 mg/l), displays a lower mean relative deviation. The testing of the efficiency of chlorine dioxide against biofilms and bacteria was made possible only after combining two new systems together, the reliable generator of chlorine dioxide, and the efficient monitor of the chlorine dioxide levels which satisfies the prescriptions of the “Drinking Water Regulation". The performed test show that under the conditions of the performed tests and with the chlorine dioxide concentration level of 0,2 mg/l, the degerming units produced by “lotronic Elektrogerätebau GmbH" and consisting of the chlorine dioxide producing unit Aquacon WH01 and the chlorine dioxidemonitoring unit Aquacon CLO2, are effective in both the inactivation bacteria in biofilms, and the prevention of formation of new biofilms.[16]

Figure 2 Stationary chlorine dioxide generator (left)and mobile chlorine dioxide analyser (right) from comp. Iotronic[17]

[16] EXNER, M., 2004: Direktor des Institutes für Hygiene und öffentliche Gesundheit der Universität Bonn [PROF. DR. MED. M. EXNER]. *Expertise. Aquacon WHO1 and Aquacon CLO2*. Internal document of lotronic Elektrogerätebau GmbH, Germany.

[17] FaltFlyerClO2KH-en. *Internal documentoflotronicProcess Monitoring, Niederlassung Deutschland.*

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Testing of the ClO_2 analyser

The Institute for technical chemistry by ZENTRUM ANGEWANDTE CHEMIE have been providing following statement regarding to the monitoring titrator Aquacon CLO2: we operated the process titrator Aquacon CLO2 (serial number: 020784) for chlorine dioxide measurement in aqueous solutions in our institute. We measured chlorine dioxide concentrations between 0.03 mg/l and 0.86 mg/l photometrically according to DIN EN 12671 (measurement of the increase of absorption at 548 nm with 1,5-bis(4-methylphenylamino-2- potassiumsulfonate)-9,10-antrachinone) and simultaneously with the Aquacon CLO2 titrator. The standard solutions of chlorine dioxide were determined by iodometric titration according to DIN EN 12671 and set as 100 percent values. A series of series of 11 measurements with each 3 different levels of concentration of chlorine dioxide were conducted. Each concentration was measured three times to ensure reproducibility (99 single measurements). The results were the following:
Average standard deviation (measurements according to DIN EN 12671): 0.0106 ppm
Average standard deviation (measurements with Aquacon CLO2): 0.0099 ppm
Average relative deviation (measurement with Aquacon and comparison 6.18 %
with iodometric titration, measuring range 0.1 — 0.3 mg/1 chlorine dioxide) Average deviation (measurement results of Aquacon CLO2 fitted with a 0.022 mg/l polynomic function and comparison with iodometric titration, measuring range 0.03 — 0.86 mg/l chlorine dioxide) In addition, 8 series of measurements with the Aquacon CLO2 were conducted by personal who received only limited instructions on how to operate the Aquacon. These measurements showed that the average standard deviation was 0.0079 mg/l. The process titrator Aquacon CLO2 (serial number: 020784) operated free of errors and functional interruptions and was very accurate as the attached results show. On the basis of the measured values can be stated that the measurement method of the company iotronic is suitable as automated procedure for the measurement of chlorine dioxide.[18]

CONCLUSION

Chlorine dioxide disinfection systems are one of the most effective ones. One of the advantages of this system comparing to chlorine disinfection of water is that there is not so number of by products of harmful substances produced from the chemical bounds of chlorine and substances in the volume of water. Study has been performed with specialists from comp. Iotronic from Germany who have many years experience with this kind of system and water analytical systems as well. They still invest to further research to make the systems more efficient and keep the trend according to dynamic of directives and norms.
Chlorine dioxide takes place also in medical, i. e. CDS, where is one of the important ingrediend. Some people from technic and chemistry arguing, that ClO2 is very toxic and agresive and therefore it should not be used for medical purpoces and to be very careful and acurate by appling on water disinfection. Advantage is, that its quite easy to produce it on site shortly before the application to the line. Continual and reliable monitoring makes this system efficient and safe. Moreover, there are a huge money saving if we do not need to heat the water up to proper temperature to kill all bacteria and viruses. There are some studies, when the chlorine dioxide was applied on farms, where the water for animals is disinfected by this kind of system and the farmers could decrease amount of antibiotica to protect them. This leads of course to more healthy meat and brings lower impact to meat consumers.

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