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## CHLORINE DECAY IN WATER DISTRIBUTION SYSTEMS

The paper presents the review of literature on chlorination as a one of frequently used water disinfection methods. A special attention is devoted to various models of chlorine decay. All of them include formulas allowing prediction of chlorine decay in the distribution systems which can prevent, to some extent, deterioration of water quality. This article is the first part of a wide research on description of contaminants' decomposition and the resulting secondary pollution in water distribution system.

### 1. INTRODUCTION

Chlorine is the most popular disinfectant used in drinking water distribution systems. In order to make the prediction of chlorine decay more true, biofilm growth and substrate utilisation more confident, numerical models have been developed.

Typically, the source water is cleared and disinfected in treatment plant before being discharged into a drinking water distribution system [20]. Disinfection is the most crucial stage in the treatment of drinking water, and chlorine (or other disinfectant) is applied in the clearwell, the final stage of treatment [1], [4], [26]. This article presents a brief review of recent studies undertaken to describe chlorine decay and disinfection by-products' formation in the system of water distribution is the first part of an extensive research on the secondary water pollution in its distribution system in the city of Lublin.

### 2. CHLORINE DEMAND IN THE NETWORK

Free chlorine is the most commonly used disinfectant because of its comparatively low price, effectiveness in killing bacteria and chemical stability in water. In the water

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distribution network, chlorine can be consumed in the bulk liquid phase, in reaction with ammonia, iron and organic compounds. Additionally chlorine disappears due to its interactions with deposits as well as due to corrosion and biomass growth on the inner pipe walls [14], [35], [36]. It is difficult to predict chlorine decay in large distribution systems, especially if they work under poor hydraulic conditions (aged pipes, small velocities, etc.) [21]. Therefore, when testing chlorine decay, it seems easier to separate the reactions associated with the bulk liquid from those associated with the pipe wall.

### 2.1. CHLORINE DEMAND OF BULK WATER

ROSSMAN et al. [23] define the bulk chlorine decay as chlorine reaction with dissolved and suspended matter, mostly natural organic matter (NOM) in the water, thus the chlorine reactions with compounds attached to or derived from pipe materials can be ignored. In most waters, the reactions of chlorine with NOM make up the majority of the chlorine demand. Chlorine also reacts with various inorganic compounds, for example, with ammonia to form different chloramines [18].

A small number of suspended particles can be present in the water distribution system because of their incomplete removal from raw water, external contamination in reservoirs or pipes, etc. [13]. They can be found in different amounts, especially in large distribution systems and they can vary throughout the system. Suspended particles affect adversely the water quality because they can carry bacteria attached to their surfaces, which protects them from disinfectant action, and they contribute to the formation of loose deposits in reservoirs and pipework. These deposits are resuspended into the water phase when the hydraulic properties of the system change (direction, velocity, water hammer, etc.) [13], [33].

Bulk decay may be isolated from wall decay by carrying out chlorine decay experiments on the source water under controlled conditions in laboratory [17]. There was found an inverse relationship between the bulk decay and an initial chlorine concentration [16].

### 2.2. CHLORINE DEMAND OF BIOFILM

In the predictions of chlorine decay, it is important to understand the relation between biofilm (biomass) formation and its chlorine demand. The deposits covering the pipe walls impede the contact of chlorine with biofilm, which consists of microorganisms like bacteria, fungi, algae and their metabolites [10], [28], [30]. The temperature above 15 °C and no flow of water in the network are optimal for the biofilm growth [25]. But LECHEVALLIER observed that the biofilm growth was possible at the temperature as low as about 6 °C [22].

WEN LU et al. [34] investigated the chlorine demand of biofilm in water distribution systems. They pointed out that the main parameters influencing biomass growth were temperature and available natural organic matter (NOM). The fraction of NOM as the source of organic carbon necessary for biofilm growth is defined as biodegradable dissolved organic carbon (BDOC). The BDOC content depends on substrate type and environmental conditions. BDOC consisting primarily of humic substances, amino acids and carbohydrates reacts with disinfectant. Amino acids, the substrate of a high demand for chlorine, resulted in a higher amount of biomass whereas carbohydrates, the substrate of a low chlorine demand, was responsible for the least biofilm mass in the presence of chlorine [5]. BUTTERFIELD [5] and WINN-JUNG [32] reported that the presence of free chlorine results in weaker biofilm growth compared to its growth in non-chlorinated water with the same quality parameters.

In practice, chlorine doses are frequently not sufficient to remove all biomass, but only do not allow the microorganisms to regrowth [8]. Adequate chlorine level in the water and the time of its contact with biofilm (the time measured from the moment of the disinfectant application to the first consumer) depend on the water quality. In Polish Standards, the sufficient dose of chlorine that prevents a biomass formation ranges from 0.3 to 0.5 of free chlorine or more if it occurs as chloramines [24]. Too high a level of free chlorine causes odour and deterioration of water quality, which has been confirmed by American and Canadian researchers [27]. Such a situation is one of the most frequent causes of consumer's complaint.

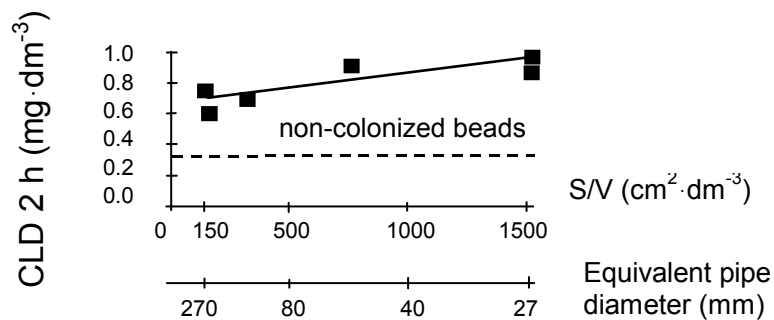


Fig. 1. Initial chlorine demand (CLD) versus  $S/V$  (at water temperature of 18 °C) [34]

Chlorine decay depends on the surface to volume ( $S/V$ ) ratio, which is an important factor in the model of chlorine decay in distribution system. The dependence is increasing when the  $S/V$  ratio varies from 150 to 1500  $\text{cm}^2/\text{dm}^3$  [34]. WEN LU et al. [34] found a linear relation between initial chlorine demand (in 2 hours) of biomass and the  $S/V$  ratio (figure 1). When the  $S/V$  increases, i.e., when a pipe diameter decreases, the

chlorine demand also increases. This means that at small pipe diameters the biomass formation should be controlled to avoid rapid chlorine consumption [34].

### 2.3. CHLORINE DEMAND OF PIPE WALL

WEN LU et al. [34] reported that chlorine decay due to the pipe wall is also affected by the surface to volume ratio ( $S/V$ ). The higher the  $S/V$  ratio, the greater the inner pipe surface which is in contact with chlorine [34]. WEN LU et al. [34] showed that chlorine consumption in new plastic pipes (PVC and PE) is negligible compared to the bulk water chlorine demand. KIÉNE et al. [11] reported that in aged grey cast-iron pipes, chlorine is principally consumed by the material corrosion and deposits [19]. Chlorine demand for different kinds of pipes is shown in figure 2.

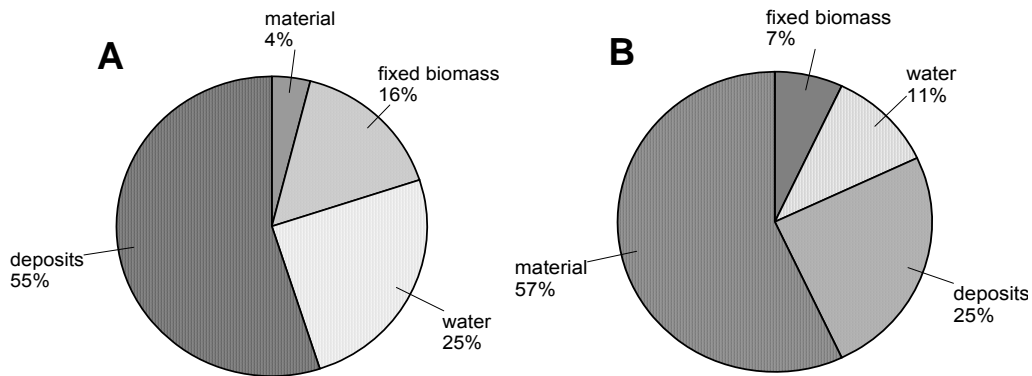


Fig. 2. Chlorine demand in 2 hours in the network made of PVC and PE pipes (a) and of grey cast-iron pipes (b) for a diameter of 250 mm [34]

As shown in figure 2, total chlorine consumption in plastic pipes in 2 hours is over two times as low as that in cast-iron pipes. In cast-iron pipes, chlorine demand of biomass becomes negligible, and chlorine is principally consumed by the material, deposits and water.

In summary, it can be concluded that the main factors influencing chlorine consumption are as follows:

- chlorine reaction with organic and inorganic chemicals in the bulk aqueous phase,
- chlorine reaction with biofilm at the pipe wall,
- chlorine consumption due to corrosion of pipe wall,
- mass transport of chlorine and other reactants between the bulk flow and the pipe wall,
- $S/V$  ratio and pipe material.

### 3. MODELLING OF CHLORINE DECAY IN WATER DISTRIBUTION SYSTEMS

Chlorine disinfection requires a constant minimum disinfectant level in all parts of a distribution system. Therefore the factors that influence chlorine decay should be identified and the models that can reliably predict chlorine residual levels in treated and distributed water should be developed. In order to achieve a balance between a chlorine dose and bacteriological quality of water, it is necessary to understand the mechanism of chlorine decay in water distribution systems and the factors affecting it. Chlorine disappears due to its reactions with compounds present in water, which in most cases remains unknown. In water distribution system, chlorine also reacts with the pipe wall. For these reasons, most kinetic models describing chlorine decay have been established empirically or semi-empirically. A basic issue of using decay equations is determination of the decay constants, which can vary with the quality of the source water, its temperature, the Reynolds number and the material properties of water pipes. Therefore, the total decay constant ( $k$ ) is often expressed by the decay due to the chlorine demand of the pipe (known as the wall decay constant ( $k_w$ )) and the decay due to the quality of water itself (known as the bulk decay constant ( $k_b$ )) [2], [3]. Bulk decay may be separated from wall decay by carrying out chlorine decay experiments on the source water under controlled conditions at the laboratory.

Within the network, dissolved chlorine reacts with NOM in the bulk water and with biofilm on the pipe walls or with the pipe wall material itself. These reactions result in a decrease in chlorine residual and a corresponding increase in disinfection by-products (DBPs), which also depend on the residence time of water in the network and the holding time in storage facilities.

The residence time of water in any distribution system changes the quality of the water, because the values of many parameters decrease or increase as a result of chemical and biological reactions which depend on the environment in the distribution system.

A number of models have been developed to predict chlorine decay in drinking water [4], [6], [7], [11], [12], [17]. Some of these models are described below.

#### 3.1. THE FIRST-ORDER CHLORINE DECAY MODEL

The most popular model is the first-order decay model in which the chlorine concentration is assumed to decay exponentially [4], [7], [17]

$$C = C_0 e^{-kt} \quad (1)$$

where  $C$  is the chlorine concentration at the time  $t$  in  $\text{mg}\cdot\text{dm}^{-3}$ ,  $C_0$  is an initial chlorine concentration in  $\text{mg}\cdot\text{dm}^{-3}$ ,  $k$  is the decay rate in  $\text{min}^{-1}$ ,  $t$  is the time in min.

The decay constant  $k$  in the model is often considered to be the bulk decay constant  $k_b$  (due to the reaction in the bulk water) and the wall decay constant  $k_w$  (due to the reaction with biofilm on the pipe wall or with the pipe wall material itself).

FANG HUA et al. [17] reported the effect of water quality parameters on the bulk decay constant of free chlorine in different water samples. They also found an empirical relationship between initial chlorine concentration and the bulk decay constant ( $k_b$ ) at a fixed temperature of three types of water. It can be represented by

$$k_b \approx \frac{0.018}{C_0} - 0.024. \quad (2)$$

As can be seen in figure 3, the decay constant  $k_b$  is inversely proportional to the initial concentration of chlorine  $C_0$ .

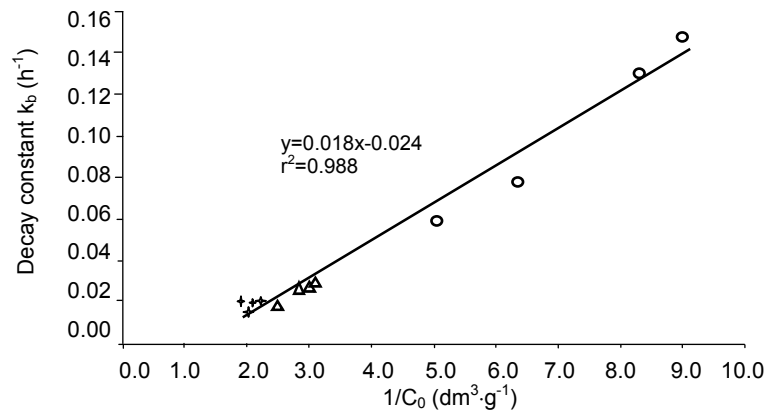


Fig. 3. The relationship between the bulk decay constant (measured at 9 °C) and the initial chlorine concentration;  $\Delta$  – final water,  $\circ$  – ambient tap water (water from a tap at the laboratory without re-chlorination, served by the same given treatment plant after flowing through the distribution pipes),  $+$  – re-chlorinated water (water from the same tap, which was re-chlorinated with sodium hypochlorite to an initial concentration of about 0.5 mg·dm<sup>3</sup>) [17]

HALLAMN et al. [16] conducted the experiment on different kinds of pipes in order to find effect of pipe material on the wall chlorine decay constant  $k_w$  in the first-order decay equation. They separated pipes into relatively reactive pipes (CI – cast iron and SI – spun iron pipes) and relatively unreactive pipes (MDPE – medium-density polyethylene, PVC – polyvinyl chloride and DICL – cement-lined ductile iron pipes).

The study aimed at examining a wider range of pipe types with repeated wall decay determinations under in situ conditions. Results of the experiment are given in figure 4.

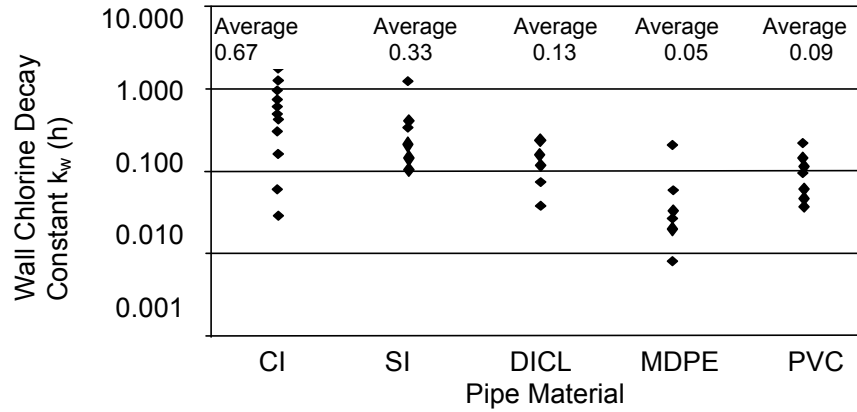


Fig. 4. Effect of pipe material on wall chlorine decay constant  $k_w$ ,  
 CI – cast iron, SI – spun iron, DIDL – cement-lined ductile iron,  
 MDPE – medium-density polyethylene, PVC – polyvinyl chloride [16]

According to the results shown in figure 4, the pipe reactivity, as judged based on the wall decay constants, was found to be as follows: CI < SI < DIDL < PVC < MDPE. Decay rates of unlined cast-iron pipes are 4–100 times as great as those of lined or plastic pipes.

### 3.2. THE SECOND-ORDER CHLORINE DECAY MODEL

CLARK [10] developed the second-order chlorine decay model based on the concept of competing reacting substances. He investigated a two-component model which accounts for both disinfectant and a fictitious reactant via the hypothetical irreversible reaction [6]



where  $A$  is the chlorine component,  $B$  is a fictitious reactive component,  $P$  is the disinfectant by-product component, and  $a$ ,  $b$  and  $p$  are the stoichiometric reaction coefficients.

Clark assumed the first-order reaction rate with respect to  $A$  and  $B$  and the second-order overall reaction rate

$$\frac{dC_A}{dt} = -k_A C_A C_B, \quad \frac{dC_B}{dt} = -k_B C_A C_B, \quad (4)$$

where  $C$  and  $k$  are the concentration and decay rate coefficient for the disinfectant (subscript  $A$ ) and a reactive component (subscript  $B$ ), respectively. The analytical so-

lution to (4) is

$$C_A(t) = \frac{C_{A,0} - aC_{B,0}/b}{1 - (aC_{B,0}/bC_{A,0}) \exp[-(bC_{A,0}/aC_{B,0} - 1)k_A C_{B,0}t]}, \quad (5)$$

where  $C_{A,0}$  and  $C_{B,0}$  are the initial concentrations of the disinfectant and a fictitious reactive component at  $t = 0$ , and the ratio of  $a$  to  $b$  and  $k_A$  are the parameters that must be estimated from experimental data. Since a reactive component is unknown, the initial concentration  $C_{B,0}$  must also be incorporated into the estimation process.

JEDAS-HECART et al. [18] and VENTRESQUE et al. [31] divided the chlorine decay into two phases, i.e., an initial phase of its immediate consumption during the first 4 hours and the second phase of its slower consumption after the first 4 hours, the latter being known as the long-term chlorine demand. Because an initial decay is rapid, DHARMARAJAH and PATANIA [9] suggested the second-order decay equation for the first phase and the first-order decay equation for the second phase.

### 3.3. COMBINED FIRST- AND SECOND-ORDER MODEL

FANG HUA et al. [17] proposed a semi-empirical combined first- and second-order model, which provides a good description of chlorine decay in the first (initial) and the second stages.

This model was derived from the equation

$$\frac{dC}{dt} = -k_1C - k_2C^2. \quad (6)$$

After integration (4) we have

$$\frac{1}{C} + \frac{k_2}{k_1} = \left( \frac{1}{C_0} + \frac{k_2}{k_1} \right) e^{k_1 t}, \quad (7)$$

where the decay constants  $k_1$  and  $k_2$  are the functions of the overall decay constant  $k$  and are determined by deriving the best fitting of equation (7) with the experimental data. The comparison of the first-order model with this combined model using experimental data for water from a treatment plant at a fixed temperature of 9 °C is shown in figure 5. It can be seen that the combined first- and second-order model provides a better description of chlorine decay than the first-order model, especially in the initial (rapid) stage.

In summary, the modelling of chlorine decay is very difficult because of complexity of water quality and its evolution in distribution system. But such modelling is important, because disinfection processes change composition and characteristics of the water distributed to a consumer. Mathematical models for the decay of chlorine allow



us to predict the formation of disinfectant by-products, which can be a health risk.

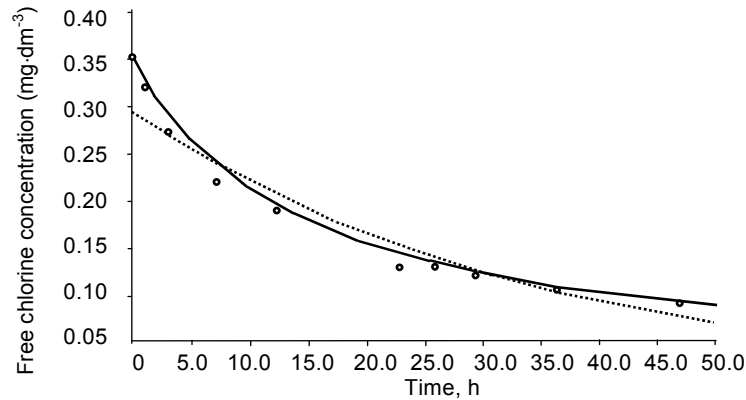


Fig. 5. Comparison of model predictions and experimental data for the water from a treatment plant: — combined first- and second-order model,  $k_1 = -0.004 \text{ h}^{-1}$ ,  $k_2 = 0.0199 \text{ dm}^{-3} \cdot \text{mg}^{-1} \cdot \text{h}^{-1}$ ,  $r^2 = 0.992$ , ..... the first-order model,  $k_b = 0.029 \text{ h}^{-1}$ ,  $r^2 = 0.926$ ,  $\circ$  experimental data [17]

Chlorine decay and its kinetics in water distribution systems depend mainly on the following factors:

- the presence of oxidizing compounds in water [20], [29],
- the presence of ammonia nitrogen and organic nitrogen (III) compounds in water [29],
- water characteristics (e.g., pH, temperature ) [20, 22, 25],
- characteristics of the distribution system (e.g., pipe material, pipe age),
- contact time (both in treatment plant and in distribution system),
- the presence of fixed and loose deposits in water.

#### 4. CONCLUSIONS

Disinfection is the most important process in treatment plant, which is to reduce the risk of infectious diseases, but the interaction between chemical disinfectants and precursor materials in source water results in the formation of disinfection by-products (DBPs). It is necessary to understand the mechanism of chlorine decay (or other disinfectant) in water distribution systems and the factors affecting it.

It is difficult to predict chlorine decay in extensive and complex distribution systems, because it reacts with the compounds present in bulk water, which in most cases remains unknown, it also reacts with the biofilm on the pipe wall or with the pipe material.

An extensive research has been conducted in attempt to characterize the nature of

reactions associated with chlorination of drinking water in distribution systems. One of the aspects of the research was the development of mathematical models for predicting the decay of chlorine and formation of DBPs themselves. Actual relationships between kinetic parameters allow a better understanding of the real effect of free chlorine on the formation of DBPs.

A large number of variables limit extensive applications of the chlorine decay models. For these reasons it is impossible to find a universal model predicting chlorine decay or formation of disinfection by-products. Generally mathematical models are established empirically or semi-empirically. Each distribution system should be considered individually, depending on its different environmental conditions.

Disinfection of drinking water and formation of disinfection by-products are still studied by many researchers who attempts to advise how to improve the water quality in municipal water treatment enterprises. So it is desirable to encourage the cooperation between researchers dealing with these problems and directors of water and sewage treatment enterprises.

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#### ROZKŁAD CHLORU W WODOCIĄGOWYCH SIECIACH DYSTRYBUCYJNYCH

Przedstawiono proces dezynfekcji wody wodociągowej za pomocą chloru, koncentrując się na przeglądzie modeli opisujących rozkład omawianego dezynfektanta. Podane zależności opisują rozkład chloru w przewodach dystrybucyjnych sieci wodociągowych, umożliwiając już na etapie modelu numerycznego analizę zawartości chloru w wodzie doprowadzanej do odbiorców. Artykuł jest wprowadzeniem do badań, które pozwalają wnikliwie analizować wtórne zanieczyszczenia wody w istniejącej sieci wodociągowej.