

Modeling of residual chlorine in water distribution system

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Abstract: Water quality within water distribution system may vary with both location and time. Water quality models are used to predict the spatial and temporal variation of water quality throughout water system. A model of residual chlorine decay in water pipe has been developed, given the consumption of chlorine in reactions with chemicals in bulk water, bio-films on pipe wall, in corrosion process, and the mass transport of chlorine from bulk water to pipe wall. Analytical methods of the flow path from water sources to the observed point and the water age of every observed node were proposed. Model is used to predict the decay of residual chlorine in an actual distribution system. Good agreement between calculated and measured values was obtained.

Keywords: residual chlorine; bulk water; corrosion; bio-film; water quality; water distribution system; flow path; water age

Introduction

Chlorine is an efficient and inexpensive disinfectant, and is widely used in drinking water works all over the world. To maintain residual chlorine in the whole water distribution system can prevent from water quality microbiological degradation. Chlorine is relatively unstable, may react with a variety of organic and inorganic compounds in bulk water or on pipe wall, and there are reactions of biofilm with chlorine. All these result is consumption of residual chlorine in distribution system.

It is very difficult to precisely predict the chlorine concentration in specific locations of the whole distribution system, because of its complexity of water distribution system, and uncertain by in reactivity of chlorine, but modeling the kinetics of residual chlorine decay in distribution system is necessary, in order to insure delivery of high quality drinking water.

The most kinetic model describing chlorine decay in water distribution system is the first order decay model in which the chlorine concentration is assumed to decay exponentially (Sharp, 1991; Hart, 1991; Clark, 1991; 1993a; 1993b; Zhang, 1992; Grayman, 1994; Chambers, 1995). The decay constant values vary with water quality, water temperature, flow rate, pipe diameter, pipe materials, the type of inner coating, pipe roughness. The decay constant values are determined by using regression method, considering these influence factors. This model can provide a fair approximation by given initial concentration and temperature. Problems of these models are too simple, some factors which control chlorine decay have not been considered, such as corrosion of pipe, bio-film on pipe wall and mass transfer of chlorine.

In other models of chlorine decay in water networks (Nakanishi, 1966; Gotoh, 1982; Biswas, 1993; Rossman, 1994; Clark, 1994; 1995; Lu, 1995), the decay constant is defined as a function of influential factors, and finds the empirical equation.

The purpose of this study is to model chlorine residual concentration in water distribution system including: (1) determination of chlorine consumption in water network; (2) analysis of the hydraulic character associated with water quality in water distribution system; (3) modeling residual chlorine concentration in water distribution system, and (4) testing the model by field data.

1 Materials and methods

Bulk decay was isolated from the other decay by carrying out chlorine decay experiments on source water under controlled conditions in laboratory. Before beginning any sampling, all the containers had to be cleaned to ensure that no chlorine demand was present. The cleaning involved washing with distilled water, standing them for 24h filled with distilled water, which had been superchlorinated (10 mg/L), flushing them with distilled water and allowing them to dry. All chlorine measurements were taken by the DPD (N, N-diethyl-*p*-phenylenediamine) colorimetric method using Toshi CRT-1000 pocket chlorine colorimeters.

2 Chlorine decay kinetics

The consumption of residual chlorine in distribution system is influenced by a number of factors including: (1) Reaction with organic and inorganic chemicals in bulk water; (2) reactions with bio-film attached to pipe wall, and (3) in corrosion process. So the chlorine decay in distribution system is:

$$\frac{dC}{dt} = -\frac{k_w C_w}{r_h} - \frac{W}{r_h} - k_b C. \quad (1)$$

Where k_w is the chlorine decay rate constant on pipe wall; k_b is the chlorine decay rate constant in bulk water; r_h is the hydraulic radius of the pipe; W is the consumption of chlorine-induced corrosion on the pipe wall; C is the chlorine concentration in bulk water; C_w is the chlorine concentration near the pipe wall.

On the equation's right, the first term is chlorine consumption in bulk water (chlorine reacts with organic and inorganic chemicals); and the second term accounts for corrosion; the third term is chlorine consumption on pipe wall.

2.1 Chlorine decay in bulk water

Two forms of chlorine are in common use as disinfectant in water, chlorine and hypo-chlorite compounds. In water, chlorine reacts very rapidly to form hypochlorous and hydrochloric acids.



$$K_i = \frac{[\text{H}^+][\text{OCl}^-]}{[\text{HOCl}]}. \quad (4)$$

The total concentrations of chlorine (Cl_2), hypochlorous acid (HOCl) and hypochlorite ion (OCl^-) is referred to as free residual chlorine. From Eq.(4), we can find that the distribution of hypochlorous acid and hypochlorite ion are affected by temperature and pH.

Chlorine can react with inorganic ions, such as Mn^{2+} , Fe^{2+} , S^{2-} . Chlorine also reacts with ammonia to form compounds called chloramines, which are slow-acting disinfectants, and are called combined chlorine. Chlorine reacts with organic matter to produce undesirable by-products, such as trihalomethanes (THM) and [MX]. Since 1979, hundreds of by-products have been identified as associated with chlorine disinfection (Krasner, 1989).

We use a generic reaction to represent all the reactions concerning with chlorine in bulk water:



Where HOCl is free residual chlorine and X is compounds reacting with chlorine. The chlorine concentration decays is:

$$\frac{d[\text{HOCl}]}{dt} = k[\text{HOCl}][X]. \quad (6)$$

Where: [] represents concentration.

It is reasonable to assume that the concentration of X is much greater than that of chlorine in bulk water, so we can take $[X]$ as a constant. We define k_b as the chlorine decay constant in bulk water, $k_b = k[X]$. This means that the reaction is pseudo-first order reaction, and the chlorine decay concentration

is:

$$\frac{d[\text{HOCl}]}{dt} = k_b[\text{HOCl}]. \tag{7}$$

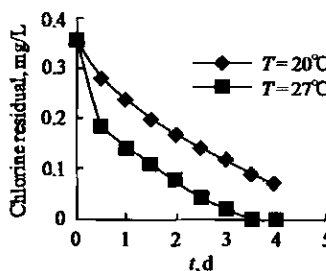
According to Arrhenius equation:

$$\frac{d \ln k_b}{dT} = \frac{E}{RT^2}. \tag{8}$$

Where: E is apparent activation energy of reaction; R is gas constant and T is absolute temperature. We can transfer (8) into:

$$\log \frac{k_{b2}}{k_{b1}} = \frac{E(T_2 - T_1)}{2.303RT_2T_1}. \tag{9}$$

If we know different k_b values at different temperatures, we can obtain the E from Eq. (9). We take treated water samples from distribution tank of the Ube City, in Japan. The characteristics of water samples are: $T = 20.7^\circ\text{C}$, $\text{pH} = 6.7$, $\text{TOC} = 2.04 \text{ mg/L}$, $[\text{HOCl}] = 0.36 \text{ mg/L}$. The water samples were placed in two clean glass bottles. During the test, the bottles were covered with laboratory film.



One bottle was placed in a low temperature incubator ($T = 20^\circ\text{C}$), the other was placed in a high temperature incubator ($T = 27^\circ\text{C}$). Twice a day, bottles were tested for free residual chlorine. Fig. 1 plotted the observed decay in free residual chlorine over time (days). In Fig. 1, the best-fit first-order decay reaction constant for these data was $k_{b20} = 0.39 \text{ day}^{-1}$. and $k_{b27} = 0.88 \text{ day}^{-1}$. We calculated activation energy according to Eq. (9), $E = 84958.2\text{J}$. Represented E by 84958.2J in Eq. (8), we can obtain chlorine decay constant (k_b) in bulk water at any temperature.

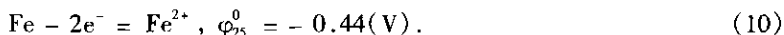
Fig. 1 The concentrations of free residual chlorine decay over time in bottle test

Table 1 summarizes the k_b values and half-life ($t_{1/2}$) values at different temperatures. The k_b depend on temperature, temperature varies from 0°C to 30°C , the k_b varies from $3.01 \times 10^{-2} \text{ day}^{-1}$ to 1.23 day^{-1} , there is a

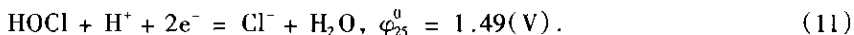
positive correlation relationship between temperature and k_b . This means that residual chlorine decreases fast, as the temperature increases.

2.2 Chlorine decay on pipe wall (corrosion)

Corrosion of water distribution pipe can play an important role in residual chlorine decay in aged metallic conduit. Corrosion of iron pipe is characterized by pitting and formation of iron oxide tubercles (Albert, 1974; Kuch, 1985; Mattsson, 1989). The presence of free residual chlorine in water promotes corrosion of ferrous metals. Electrochemical corrosion can be viewed in terms of oxidation and reduction reactions. From Eq. (4), we know that Cl_2 is not present at the pH of normal water, and hypochlorous acid (HOCl) is dominant over hypochlorite ion (OCl^-) in water under $\text{pH} 7.5$. In tap water, $\text{PH} \approx 7.5$, so the compound undergoing reduction reaction is hypochlorous acid (HOCl). For corrosion to occur, at the anode, the oxidation of ferrous metals occurs:



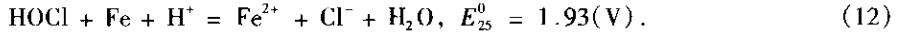
At the cathode, reduction reaction occurs:



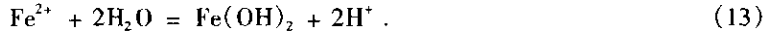
From Eq. (10) and Eq. (11), we find the whole electrochemical corrosion equation:

Table 1 The bulk decay coefficient and half-life of chlorine

Temperature, $^\circ\text{C}$	k_b, d^{-1}	$T_{1/2}, \text{d}$
0	3.01×10^{-2}	22.88
5	5.94×10^{-2}	11.69
10	1.14×10^{-1}	6.09
15	2.13×10^{-1}	3.26
20	3.90×10^{-1}	1.78
25	7.00×10^{-1}	0.99
30	1.23	0.56



Fe^{2+} is the corrosion product, pit contains high concentration of FeCl_2 . Hydrolysis of FeCl_2 can occur:



Both hydrogen and chlorides stimulate the dissolution of iron, the corrosion process accelerates with time, and the entire corrosion process has the property of "self-catalysis". The electromotive force of (12) is:

$$E = E^0 - \frac{RT}{zF} \ln \frac{[\text{Cl}^-][\text{Fe}^{2+}]}{[\text{HOCl}]}. \quad (14)$$

Where E is the electromotive force of corrosion reaction; E^0 is the standard electromotive force at temperature 25°C . In this reaction $E^0 = 1.93\text{V}$; R is the gas constant, $R = 8.314 \text{ J}/(\text{mol} \cdot \text{K})$; F is Faraday constant, $F = 96485\text{C}$; z is the number of electrons transferred in reaction, and $z = 10$; T is the absolute temperature (K); the brackets denote the activities of the HOCl. From the form of (14), we find the $[\text{HOCl}]$:

$$\log[\text{HOCl}] = \frac{zF(E - E^0)}{2.303 \times RT} = \frac{10078.26}{T}(E - E^0). \quad (15)$$

Eq.(15) can be used to determine the consumption of chlorine in the corrosion process.

The value of E varies with different pipe materials, age and surface roughness. For a definite distribution pipe, the E is a constant. Thus we can find the consumption of chlorine in the corrosion process on distribution pipe wall. We define W as chlorine demand caused by pipe wall:

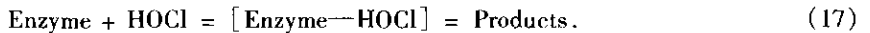
$$W = \log[\text{HOCl}] = \frac{10078.26(E - E^0)}{T} = \frac{k_c}{T}. \quad (16)$$

Where k_c is the corrosion coefficient of chlorine decay and T is the absolute temperature(K).

2.3 Chlorine decay on pipe wall(reacting with biofilm and tubercles)

The formations of tubercles and bio-film on pipe wall have been found in drinking water distribution system in many investigations, tubercles and bio-film can result in a significant loss of free residual chlorine over time (Tuovinen, 1980; Kirmyer, 1983; Lechevallier, 1990; 1993; 1996; Block, 1993; Chen, 1996; Dukan, 1996). Tubercles contain several inorganisms and organisms and various heterotrophic microoganisms. In fact, hypochlorous acid(HOCl) has much higher bactericidal efficiency than that of hypochlorite ion(OCl^-). Because hypochlorous acid(HOCl) is a molecule without charges, it can diffuse the surface of bacteria with negative charge, and penetrate the biofilm and inactivate attached microorganisms. But hypochlorite ion(OCl^-) with negative charge is difficult to access the bacteria with negative charge. As we know, at the pH of normal tap water, hypochlorous acid(HOCl) is dominant over hypochlorite ion(OCl^-), so the consumption of chlorine reacting with bacteria may play a dominant role in overall free residual chlorine decay in water distribution system.

We use a generic reaction to represent all reactions concerning with chlorine on the pipe wall:



The chlorine concentration decay is:

$$\frac{d[\text{HOCl}]}{dt} = k[\text{HOCl}][\text{Enzyme}]. \quad (18)$$

Where $[\]$ represents concentration; HOCl is the free residual chlorine; enzyme is all tubercles and bio-film reacting with chlorine on the pipe wall.

It is also reasonable to assume that the concentration of enzyme is much greater than that of chlorine on pipe wall, so we can take $[\text{Enzyme}]$ as a constant. We define k_w as the chlorine decay constant on pipe wall, $k_w = k[\text{Enzyme}]$. This means that the reaction is pseudo-first order reaction, and the chlorine decay concentration on pipe wall is

$$\frac{dC}{dt} = \frac{d[\text{HOCl}]}{dt} = k_w[\text{HOCl}] = k_w C_w. \quad (19)$$

Where k_w is the chlorine decay constant on the pipe wall and C_w is the concentration of free residual chlorine on the pipe wall.

There is a laminar flow film in the boundary layer region of pipe wall. From Fick's law, in the laminar film, the mass transfer velocity of free residual chlorine is:

$$V_N = k_f(C - C_w). \quad (20)$$

Where k_f is the mass transfer coefficient; C is the concentration of free residual chlorine in bulk water; C_w is the concentration of free residual chlorine on the pipe wall.

If the reaction of chlorine consumption on the pipe wall is controlled by the chlorine diffusion from bulk flow to the pipe wall, there is no chlorine accumulation on the pipe wall, so $(C - C_w) \approx C$, and:

$$V_N \approx k_f C. \quad (21)$$

In this situation, the rate of chlorine consumption on the pipe wall is the same rate of chlorine transportation from bulk flow to the pipe wall. Then we find:

$$k_w C_w = k_f C. \quad (22)$$

The mass transfer coefficient is usually expressed by using Sherwood number as follows (Rossman, 1994; Clark, 1995; Edwards, 1976; Kays, 1993):

$$k_f = Sh \frac{D}{d}. \quad (23)$$

Where Sh is the Sherwood number; D is the molecular diffusivity of chlorine in water; d is the pipe diameter. In the laminar flow film, the Sherwood number is given (Edwards, 1976):

$$Sh = 3.65 + \frac{0.0668(d/L) Re \times Sc}{1 + 0.04[(d/L) Re \times Sc]^{2/3}}. \quad (24)$$

Where Re is the Reynolds number and Sc is the Schmidt number.

The values of Re and Sc are given:

$$Re = \frac{ud}{\nu}, \quad (25)$$

$$Sc = \frac{\nu}{D}. \quad (26)$$

Where ν is the kinematic viscosity of water.

Use these relationships, the k_f can be written as:

$$k_f = \left[3.65 + \frac{0.0668 \left(\frac{ud^2}{LD} \right)}{1 + 0.04 \left(\frac{ud^2}{LD} \right)^{2/3}} \right] \frac{D}{d}. \quad (27)$$

From Eq. (27), we find that the value of k_b is a function of flow velocity, pipe length, pipe diameter, and temperature (the diffusivity vary with temperature).

3 Chlorine decay in single pipe

According to the above relationships and under steady-state condition, we consider Eq. (1), and find the overall chlorine decay in a single pipe as follows:

$$\frac{dC}{dt} = - \left(\frac{k_f}{r_h} + k_b \right) C - \frac{10^{k_c/T}}{r_h}. \quad (28)$$

The overall decay of free residual chlorine in water pipe is a function of flow velocity, pipe length, pipe diameter, pipe materials, pipe age and pipe roughness, temperature, and reaction activation energy.

We measured the concentration of residual chlorine over time (hours) new and old cast iron pipes of

different diameters. We plotted the $\ln(C/C_0)$ over time and found the slope(h^{-1}). The results are shown in Table 2. From Table 2, we find that the rate of residual chlorine decay increases with the pipe diameter decreasing, and the rate of residual chlorine decay increases from new cast iron pipe to old cast iron pipe.

We can calculate the value of k_b from Eq.(8), the k_f from Eq.(27), and k_c from Eq.(16). For a single drinking water pipe, we may find free residual chlorine decay:

$$\frac{dC}{dt} = -KC - A. \tag{29}$$

$$K = \frac{k_f}{r_h} + k_b. \tag{30}$$

$$A = \frac{10^{k_c/T}}{r_h}. \tag{31}$$

$$C_t = \left(C_0 + \frac{A}{K} \right) e^{-Kt} - \frac{A}{K}. \tag{32}$$

Where C_t is the concentration of chlorine at any time; C_0 is the initial concentration of chlorine.

For a relatively new lining drinking water pipe, we neglect the consumption of residual chlorine in corrosion process, we can find free residual chlorine decay:

$$\frac{dC}{dt} = -\left(\frac{k_f}{r_h} + k_b\right)C = KC, \tag{33}$$

$$C_t = C_0 e^{-Kt}. \tag{34}$$

4 Analysis of flow paths in a network

The loop network is different from single pipe and parallel pipe, and the number of paths water flow from water sources to the observed point is concerned with the amount of loop. The network consists of 14 pipes as in Fig.2. The water flow from 1(start point) to 10(end point) pass through 8 paths. The analytic method of the water paths in the loop network is shown in Fig.3

Make the variable *MP* of the network to quadratic matrix. The input and output are marked as starting point and ending point, and then we change the network into a tree-like structure and find the paths. In addition, when there are paths, the *MP* is marked as one and there is no any path the *MP* is zero. According to the above method, the amount of branch can be determined.

The analytical algorithm is as follows (Fig. 4):
 (1) The number of the start point and the end point is determined first and input the datum analyzed;
 (2) determine the start point and the end point of a network

Table 2 At $T=7^{\circ}C$, the slope values(h^{-1}) of $\ln(C/C_0) - t$

	DN100	DN150	DN200	DN250
S_{new}	3.30E-01	1.50E-01	1.30E-01	8.30E-02
S_{old}	4.50E-01	2.60E-01	1.50E-01	

Notes: S_{new} is slope of new cast pipe, S_{old} is slope of old pipe

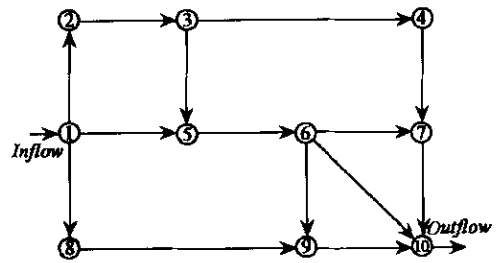


Fig.2 The network consists of 14 pipes

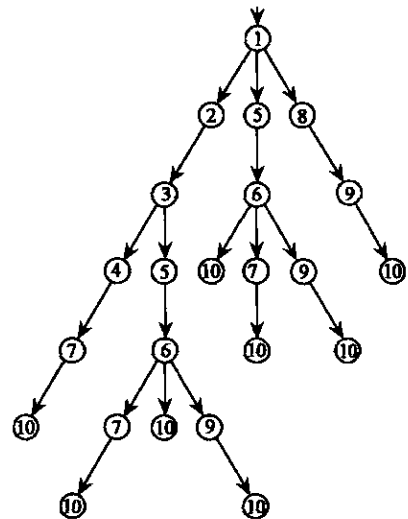


Fig.3 Analysis of flow path

and choose essential factor of flow from start to end, and then find the amount of branch in every nodes; (3) find the next nodes forward sequentially; (4) decide every node by the different from amount of branch and examine the nodes for finding the paths; (5) the datum from (1) to (4) should be calculated repeatedly. The datum needs to be calculated once more, until the path reaches the end.

The data should be marked with amount of paths, number of every path and node. In Fig. 3, all the symbols are presented as follows: $MP(i, j)$: variable of path; $MP(\text{input}, \text{output})$; K : times of calculation; f : number of branch; $PS(K_1)$: the start point; $PS(K_2)$: the end point; $B(f)$: the amount of branch.

5 Water age

In water distribution system, the travel time of water from source to a particular node is called water age. It is an important parameter to quantify the variation of water quality.

The water age may be calculated by the following formulas:

$$t = \frac{L}{v} \tag{35}$$

$$v = \frac{4q}{\pi D^2} \tag{36}$$

$$t = \frac{\pi D^2 L}{4q} \tag{37}$$

Where D is the pipe diameter, L is the pipe length, q is the pipe flow and v is the velocity.

Hazen-Williams equation is used to set up the relationship between t and h (head loss):

$$t = 2.08 C^{-0.76} q^{-0.24} L^{1.41} h^{-0.41} \text{ (m/s)}. \tag{38}$$

In Fig. 5, the water age at point C is given:

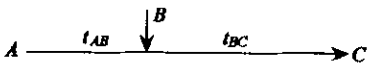


Fig. 5 A single pipe

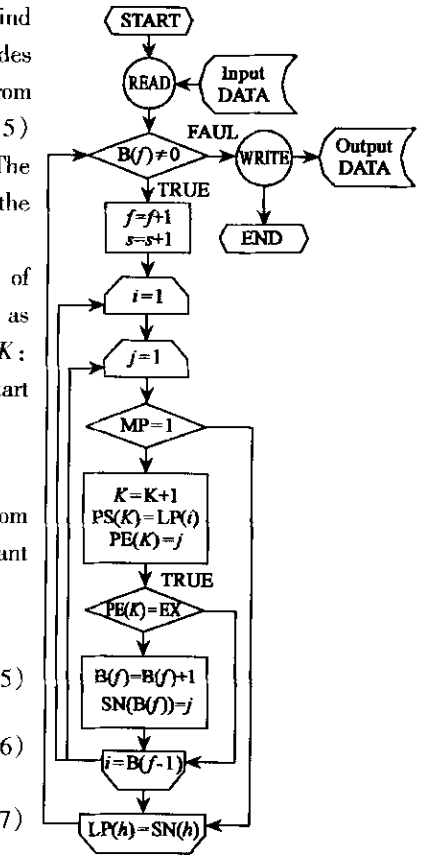


Fig. 4 Scheme of analytical algorithm for the flow path

$$t_c = t_{AB} + t_{BC} \tag{39}$$

In Fig. 6, the water age at point B is given:

$$t_b = \frac{q_1 t_1 + q_2 t_2}{q_1 + q_2} \tag{40}$$

In loop network, the water age at a specified node may be defined as:

$$t_i = \frac{\sum_{j=1}^n q_j t_{ij}}{Q_i} \tag{41}$$

Where t_i is the water age of point i ; n is the road number of water transfer from start point to node i ; q_j is the flow to point i on free water road; t_{ij} is the retention time to point i on the free water road; Q_i is the outflow at point i .

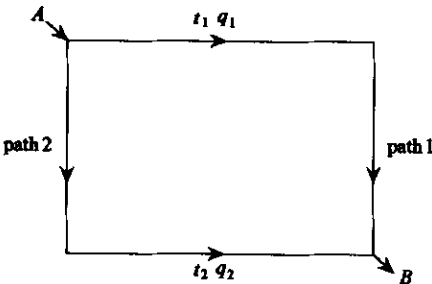


Fig. 6 Parallel pipe

6 Application in field test

The model of residual chlorine decay was tested on one

of the water distribution systems of Ube City in Japan. The service area had 40000 residential, average water use was 21000 m³/d. The water came from a water distribution tank, which had capacity of 8000 m³/d. The distribution system was composed of pipe diameter from 0.075m to 0.7m, and the total length of pipe was 49 km. The pipes were cement-mortar lining ductile iron pipes. Sampling was conducted at nine locations throughout the service area. Fig.7 illustrates the water distribution system subjected for our study and the flow directions are obtained by our analytic method. Table 3 shows the number of paths and the water age of each observation point.

In Fig. 8, one may find that the field data is lower than the calculated results at point two and point five. We think that there may be a dead-end section. In these dead-end sections, the water will have a very long residence time and the chlorine residual decay more quickly than that predicted by model. Or, these sections contain pipes that are old age, the corrosion consumption of chlorine residual on pipe wall is probably significant. At point six, the error may be attributed to the instrument error used for measuring residual chlorine. At other sampling locations, there is good agreement between the calculated results and the measured values.

Table 3 Water age and number of paths

Observation point	Water age, h	Number of path
01	Starting point	Starting point
02	3.98	4
03	3.70	1
04	5.68	5
05	3.97	1
06	7.90	2
07	9.05	2
08	10.82	15
09	11.38	6

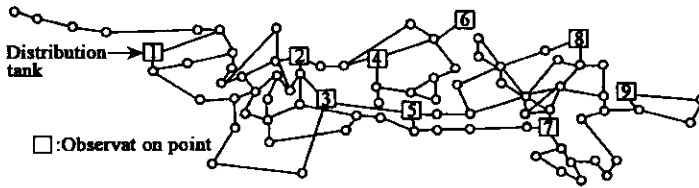


Fig.7 Flow directions and observation point in objective water distribution system

The comparison between calculated results and the measured values means that this model of chlorine residual decay in the water distribution system is considered to be available.

7 Conclusions

In order to ensure delivery of high quality drinking water from treatment plants to consumers, maintaining adequate levels of residual chlorine is very important, so it is necessary to study and model the kinetics of residual chlorine decay in distribution system. The accurate prediction model of residual chlorine must be based on the mechanisms of chlorine decay in water distribution system. In this study, we consider the consumption of chlorine reacting with chemicals in bulk water, the reaction of chlorine with biofilm on the pipe wall, the consumption of chlorine in the corrosion process, and the mass transport of chlorine from bulk water to pipe wall. The residual chlorine loss is influenced by flow velocity, pipe length, pipe diameter, pipe materials, pipe age and pipe roughness, temperature, and reaction activation energy.

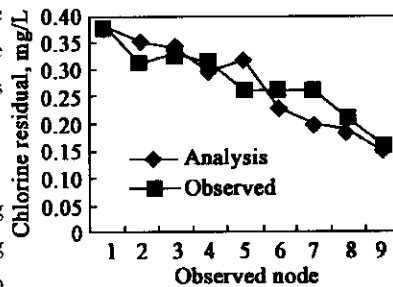


Fig.8 Comparison of observant and calculate values of chlorine residual at nine monitor point

We have analyzed the hydraulic character of water distribution system, and given the computer

algorithm of water network analysis. The concentration of free chlorine residual and retention time in each pipe are proposed. The model is used to predict the decay of chlorine residual in an actual distribution system. It is found that free chlorine residual varies both spatially and temporally. There is good agreement between calculated and measured values. Because the water quality of treated water may deteriorate in distribution system, the model of chlorine residual decay in water network is a useful tool for water utilization and engineering.

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