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Influences of pipe materials and hydraulic conditions on the process of trihalomethanes formation in water distribution network

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ABSTRACT

In this paper, the influences of different pipe materials and hydraulic conditions on the formation process of trihalomethanes (THMs) were investigated. Water used in this research was taken from a Chinese water treatment plant in the northeastern part of China. The results showed that, under the same condition, the ranking of THM concentration in different pipe materials was: PVC > stainless steel > ductile iron, while it was different for chlorine decay. The variation of THMs in distribution system simulator is consistent with that of bulk water experiment. THM amount is only slightly higher in water distribution system simulator. The trend is not as distinct as that of chlorine decay. A higher flow velocity of water is associated with higher THM concentration, but it was not obvious compared with the decay of chlorine. For the same distance, different flow velocities and diameters, THM concentration was dependent on the influential degree of retention time and flow velocity. For the same distance and different flow velocities, THM concentration increased with the increase of pipe diameter. THM levels in real water distribution networks are dependent on various influences.

Keywords: Water distribution network; Trihalomethanes (THMs); Pipe material; Hydraulic condition

1. Introduction

Chlorine has been widely used in drinking water disinfection around the world because of its advantages such as low price, good disinfection effectiveness and convenience. However, when chlorine reacts with organic components such as fulvic and humic acids, disinfection by-products (DBPs) were formed [1,2]. Traditionally, trihalomethanes (THMs) have been the research focus because of their chemical properties, toxicology and health risks [3–6]. Consequently, the effective management of chlorine at water treatment works and in distribution systems is of great importance in ensuring that water supplied to customers is safe with minimum acceptable risk from bacteriological and chemical impurities [1,10].

Villanueva et al. found that THMs and haloacetic acids (HAAs) varied considerably between different drinking waters according to their water sources. They also found that despite these differences, total and specific HAA species show a high correlation with total and specific THM compounds. Thus, HAA levels could be predicted from THM levels [7]. Jean-B

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et al. showed that seasonal variations in THM were mainly associated with variations in organic precursors and to changes in water temperature in treated waters of the three major drinking water utilities of the Quebec City area. Also, initial THM formation (in finished waters leaving the plant) was a good predictor of THM levels at distribution system extremities [8,9]. H. Pourmoghaddas et al. suggested that the percentage of total organic halogens made up of total THMs plus total HAAs significantly increases with increasing bromide ion concentrations and pH [10]. Pingping et al. showed that total organic carbon (TOC) and UV-254 had a poor correlation with THM precursors; in the range of pH 5–10, THM increased with the increase of pH value; THMs levels positively correlated with chlorine dosage, reaction time and temperature [11]. Danhong et al. investigated the influences of reaction time, organic matter, temperature, pH value, ammonia concentration and bromide ion on THMs formation, and the results showed that all influences except ammonia would promote THM formation [12].

So far, besides investigations of formation and determination of THMs, previous research mainly focused on models predicting formation of THMs in drinking water, influences on THM formation during water treatment processes and variations of THM concentrations etc. [13–15]. The research on THM formation and variation in water distribution networks is still under the preliminary stage, and there is little investigation on pipe materials and hydraulic conditions. However, it is important for the research of DBPs and water quality modelling in real networks. In this study, experimental scheme of static pipe sections and dynamic simulation was built, and the influences of pipe materials and hydraulic conditions on the formation of THMs were investigated.

2. Materials and methods

2.1. Experimental equipment

2.1.1. Static pipe section experiment

Disinfectant reacts with sedimentation, biofilm, erosion tubercles and pipe wall. In order to know influences of the reactions on the formation of THMs, static pipe section experiment was done. Static reactors were made of different pipe sections with various material and age cut from real network. The reactor is shown in Fig. 1. The pipe materials of reactors are ductile iron without lining, stainless steel and PVC respectively, and the diameter is 80 mm. Coupon is set in reactors.

Fig. 1. Static pipe section reactor.

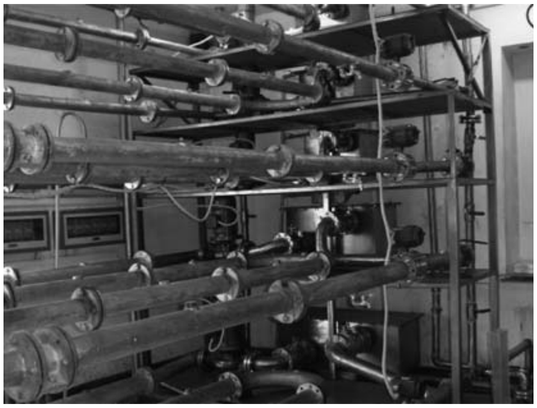

Fig. 2. Picture of distribution system simulator.

2.1.2. Dynamic simulation experiment

In this study, dynamic distribution system simulator was used as the reaction equipment, as shown in Fig. 2.

Circularity distribution system is made up of five layers of circularity pipelines, as shown in Fig. 3. From the bottom up, diameters of the first and second layers are 100 and 80 mm respectively, and the diameter of the top three layers is 50 mm. The whole distribution system simulator is made of ductile iron pipeline without lining, and the length of each layer is 17.5 m. There is distinct erosion tubercles and biofilm inside the pipe. Water used for the distribution system simulator can be supplied by water tank of each layer, and one-layer circulation can be carried out by variable frequency speed governing pumps. Water can also be supplied by elevated water tank, and combination circulation can be carried out by the larger variable frequency speed governing pump of the lower layer. The length of pipeline can be changed, and pipelines of different diameters can be combined, meeting different experimental requirements.

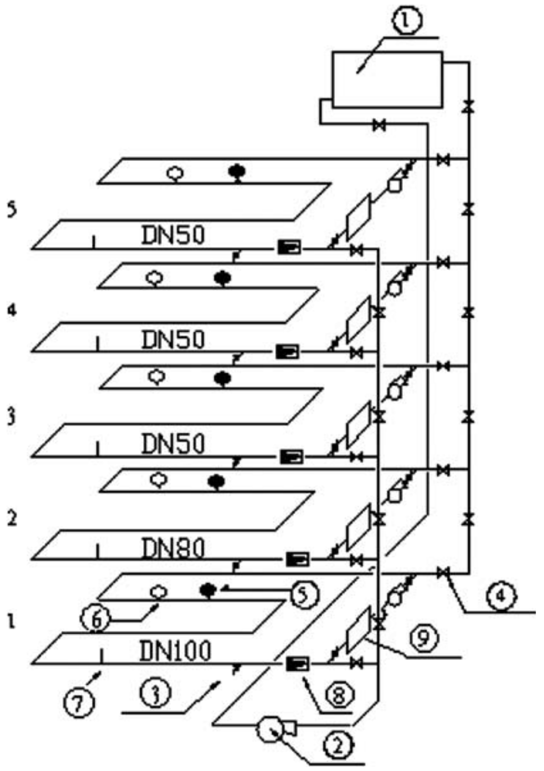

Fig. 3. Schematic diagram of distribution system simulator (1) elevated water tank, (2) circulating pump and lift pump, (3) sampling place, (4) valve, (5) temperature sensor, (6) pressure gauge, (7) coupon, (8) flowmeter, and (9) circulation water tank.

2.2. Experiment methods

2.2.1. Static pipe section experiment

Filtered water (without chlorination) from the water treatment plant was used as experimental water. TOC and pH values of the filtered water are 3.16 mg/L and 6.57 respectively.

Sodium hypochlorite was added into filtered water and pH value was adjusted to 7.3 ± 0.3. After simulating reaction period for 2 h in a clear water tank, the sampling water was filled into three static pipe sections, and kept at room temperature. Chlorine and THM measurements were taken when the reaction time was 0, 2, 4, 6, 8, 10, 12 and 24 h.

2.2.2. Dynamic simulation experiment

Tap water was used as experimental water as dynamic experiment consumes too much water. TOC and pH values of tap water are 5.20 and 6.5 mg/L respectively. Before experiment, the water was kept open for 24 h to remove chlorine residual.

(1) The formation of THMs under the same diameter, the same retention time and different flow velocities.

30 mL of concentrated sodium hypochlorite was added into 500 L-experimental water of the simulator, and a large circulating pump was used so that chlorine in water can be mixed throughly by the circulation of riser pipe. 2 h later, chlorine concentration and pH value were 1.41 and 7.33 mg/L respectively. Water was injected into each layer with the diameter of 50 mm. and the flow velocity was adjusted to 0.3, 0.6, and 0.9 m/s respectively. Chlorine and THM measurements of the sampling water after the same retention time were taken Parallel test of bulk water was done simultaneously.

(2) The formation of THMs under the same flowing distance and different flow velocities.

In real networks, if the distance of water from water treatment plant to specific user is fixed, namely the distance is fixed, then it changed flow velocity, retention time and pipe diameter. According to that fact, the flowing distance was set as 3,500 m in the dynamic experiment. The initial chlorine and pH values were 1.41 and 7.23 mg/L respectively, and the flow velocity of each layer was set to 0.3, 0.6, and 0.9 m/s. Thus, the time through the same distance is 194.45 min (0.3 m/s), 97.22 min (0.6 m/s), and 64.15 min (0.9 m/s). THM measurements was taken each time.

2.3. Analyses of parameters and methods

Conventional parameters were measured according to the national standard method [16]. Chlorine measurements were taken by the N,N-diethyl-p-phenylenediamine colorimetric method. Liquid-liquid extraction and gas chromatography with electron capture detector were used to confirm THM concentration. Chlorine measurements were taken in triplicate using one Hach (Chlorine Pocket Colorimeter II) and either one or two Hanna (HI-95711) chlorine colorimeters, to improve the accuracy of the measurements. THMs measurements were also taken in triplicate.

3. Results and discussion

3.1. Pipe material

A number of investigators have conducted research into the chlorine decay modelling, including Powell et al. [17], Hua et al. [18], and Rossman et al. [19]. Clark et al. [20] reviewed current and historical research efforts related to chlorine decay modelling [20]. They also present a mathematical model based on the use of two second-order terms for predicting chlorine decay.

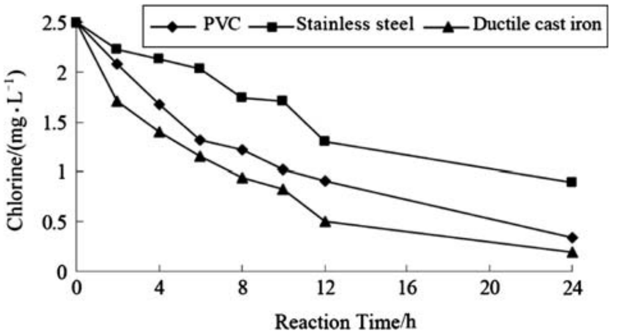

Fig. 4. Influence of pipe material on residual chlorine decay.

Table 1
Standard deviation of k for each pipe material

Pipe materials	k (h^{-1})	Standard deviation
Ductile iron	0.1	0.0024
PVC	0.08	0.0021
Stainless steel	0.04	0.00044

It is widely accepted that a large amount of chlorine is consumed rapidly in the first few minutes. Some results showed that: up to 37–53% of the total amount of free chlorine consumed in the 4 h test period is being consumed in the first 5 min [21].

First-order decay model is simple to apply, especially for network modelling purposes although it has some limitations. So in this study, for the simulation of chlorine decay, we choose first-order model. The chlorine decay coefficients of ductile iron, PVC, and stainless steel are: 0.1, 0.08, and 0.04 h^{-1} respectively. The results showed that the ranking of chlorine decay velocity in different pipe materials was ductile iron > PVC > stainless steel. Chlorine decayed faster during the first 12 h, and much slower in the last 12 h, as shown in Fig. 4.

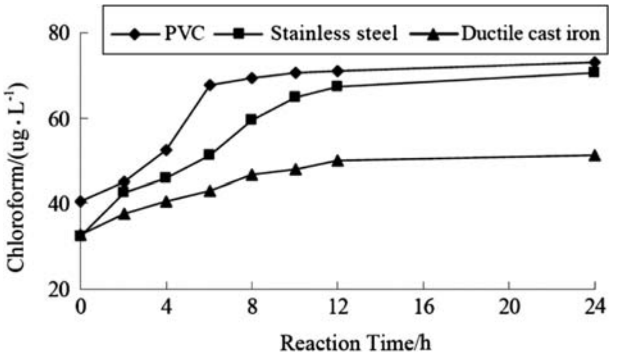

Fig. 5. Influence of pipe material on chloroform production.

The standard deviation of k for each pipe material is shown in Table 1.

Influence of pipe materials on chloroform formation is shown in Fig. 5. During the first 12 h, formation velocity of chloroform was faster, while that during the period of 12–24 h was much slower, which was corresponding to chlorine decay. However, the ranking of chloroform concentration in different pipe materials was PVC > stainless steel > ductile iron, which was different from the ranking of chlorine decay.

In order to find out the reason for the phenomena mentioned above, electron microscope scanning of the

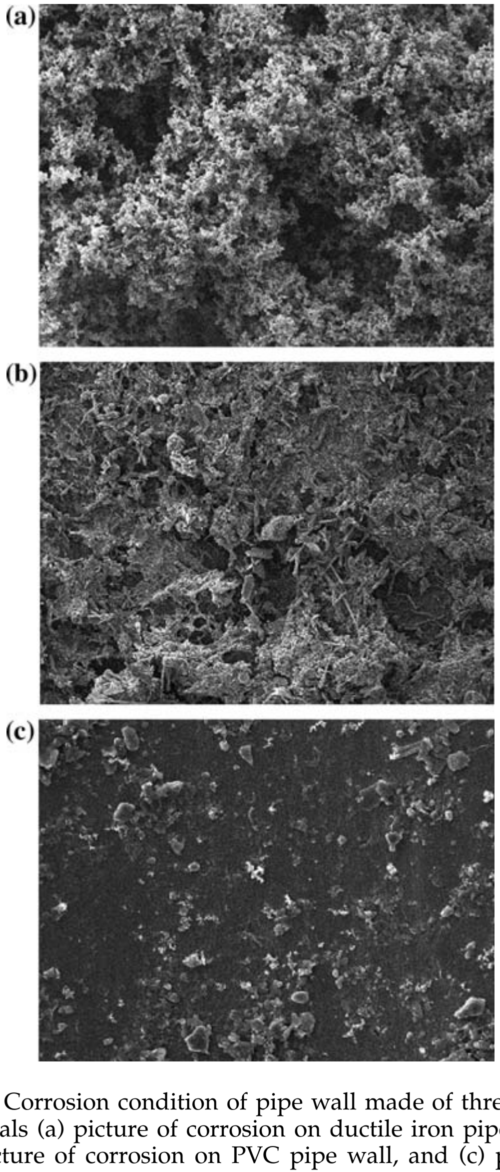

Fig. 6. Corrosion condition of pipe wall made of three pipe materials (a) picture of corrosion on ductile iron pipe wall, (b) picture of corrosion on PVC pipe wall, and (c) picture of corrosion on stainless steel pipe wall.

coupon on three pipe materials was taken. The photographs are shown in Fig. 6, and the magnification was 500× .

Ductile iron: It can be seen from Fig. 6(a) that serious corrosion occurred inside the pipe section, and lots of sedimentations attached to the pipe wall, both of which mixed together. Various corrosion products contacted loosely, and it was easy to fall off. Moreover, much space existed, and tubercles formed. This is because ductile iron contains a certain amount of carbon impurities, whose electrochemical potential is different from that of iron substrate. Lots of corrosion micro-batteries form, leading to electrochemical corrosion. As a result, roughness inside the pipe wall increases, providing a haven for microorganisms, especially for iron bacteria which can adhere to the bottom of corrosion pits inside cast iron pipe walls. Iron bacteria absorb ferrous ion in water and massive ferric hydroxide is generated, leading to the formation of tubercles on pipe walls and pipeline will be plugged. The area with tubercles becomes anaerobic zone and sulphate reducing bacteria will be produced and increased. Thus, under the function of hydrogenase, local corrosion will occur on cast iron under tubercles. Therefore, erosion of cast iron pipelines is the most serious, and chlorine decay in cast iron pipelines is the most remarkable compared with the other two materials. However, THM levels are the least. This is because most of free chlorine reacts with tubercles and biofilms, and only a little part of that reacts with precursors in water to form THMs.

PVC: Fig. 6(b) shows that little corrosion exists, but lots of scales deposit on pipe walls. The main components of them are post-precipitation of pipelines. The deposits are easy to be broken off and microorganisms can grow up in the great space between deposits. Therefore, corrosion micro-battery is unable to form on PVC pipe with the main component of polyvinylchloride, and it is not easy to be corroded. However, compared with stainless steel, PVC is rougher, which also increases chlorine consumption. Although bacteria and deposits attached on pipe wall consume part of chlorine residual, the deposits and dissolved biodegradable organics also provide DBP precursors for chlorination reaction. In previous research, biofilm activity in PVC is the highest in the pipe materials being studied [22]. Accordingly, chloroform in PVC pipes is higher than that in stainless steel and cast iron pipes.

Stainless steel: As shown in Fig. 6(c), little corrosion and post-precipitation exist. This suggests that stainless steel is not easy to be corroded and its pipe wall goes against adhesion of deposits. As 99% of stainless steel pipe is iron, its surface is electrochemically uniform, and there is no corrosion micro-battery. Moreover, as the pipe wall is smooth, little deposit exists. Thus, there is almost no corrosion on stainless steel pipes. As a result, chlorine consumption is the least, and most part of chlorine reacts with precursors and THM generate.

Previous results showed that, THM formation depends on numerous factors including chlorine dose, retention times in unit processes and pipes, temperature, raw water quality and changes in the water chemistry [23].

3.2. Hydraulic conditions

3.2.1. The formation of THMs under the same diameter, the same retention time and different flow velocities

Decay of residual chlorine in different flow velocities is shown in Fig. 7. It can be seen from the comparison between bulk water experiment and dynamic simulation experiment that chlorine decays much faster in networks than in bulk water.

The chlorine decay coefficients under the flow velocities of 0.3, 0.6 and 0.9 m/s are 0.30, 0.59, and 0.74 h^{-1} respectively. Chlorine decay rate increases with the increase of velocity. This is because when water flows inside pipes, most chlorine reacts with pipe wall. As flow velocity increases, oxygen accelerates corrosion reaction inside pipe wall, which not only aggravates erosion, but also increases chlorine consumption.

The standard deviation of k for each flow velocity is shown in Table 2.

Fig. 8 shows that during the same retention time, the variation of THM amount in distribution system simulator is consistent with that of bulk water experiment. THM amount is only slightly higher in distribution system simulator, not as distinct as chlorine

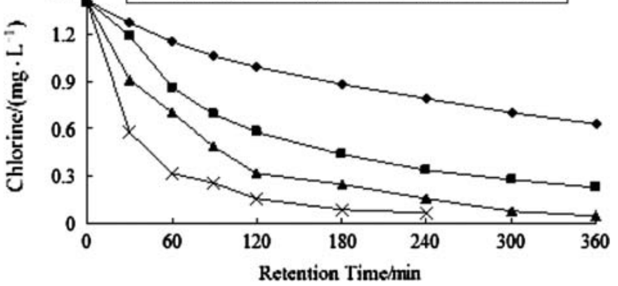

Fig. 7. Residual chlorine decay curves in different flow velocities.

Table 2
Standard deviation of k for each flow velocity

Flow velocity (m/s)	k (h^{-1})	Standard deviation
0.3	0.30	0.0045
0.6	0.59	0.021
0.9	0.74	0.019

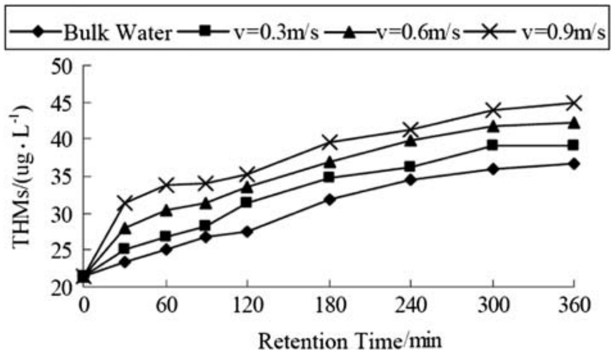

Fig. 8. THMs production curve in different flow velocities.

decay (Fig. 7). This may be because scales and tubercles adhere to pipe walls, and biofilm on pipe wall contains some THM precursors. So that in dynamic experiment, the substances react with chlorine as water flows, and THM forms. The increase of THMs reduces the difference of that between bulk water and distribution system simulator.

As shown in Fig. 8, THMs amount increases with the increase of flow velocity, but it is not as distinct as chlorine decay. One of the reasons is that chlorine mixes and reacts with THM precursors more thoroughly as flow velocity increases, accelerating THM formation. The other reason is although chlorine is one of the reactants to create THMs, most of chlorine reacts with pipe wall. Therefore, although the increase of velocity greatly increases chlorine decay, there is no distinct influence on THM formation.

It is obvious that chlorine is an important condition for THM formation, but not unique. THMs level in real networks is dependent on various influences.

3.2.2. The formation of THMs under the same distance, different flow velocities and diameters

As shown in Fig. 9, THMs' level is significantly higher in water with a flow velocity of 0.3 m/s than 0.6 m/s and 0.9 m/s. The retention time of water with a lower velocity is longer than that of a higher velocity for the same flow distance. THMs' level increases as the increase of retention time. As analysed in

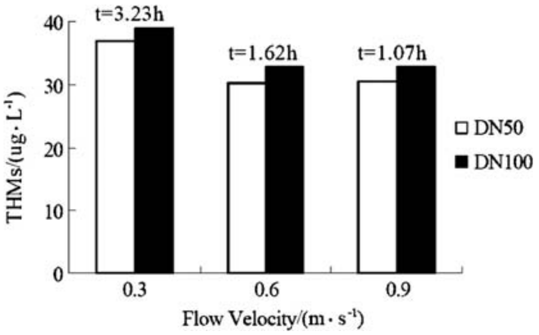

Fig. 9. Histogram of THM production in the same distance and different velocities.

Section 3.2.1, THM formation increases with the increase of velocity, and the influence is not significant. It is reasonable to conclude that when both retention time and flow velocity influence THM formation, retention time plays a dominant role. Retention time of water with flow velocity of 0.6 m/s is slightly higher than 0.9 m/s. So influence of retention time on THM formation counteracts that of velocity. THM levels of the two conditions are close to each other as shown in Fig. 9. The results suggest that when both retention time and velocity influence THM formation, its variation is decided by influential degree of the two effects.

To explain the effect of diameter on THM formation, contact rate is introduced in advance. Contact rate is defined as contact area with pipe walls of water per unit volume in pipelines. Then contact rate decreases with the increase of diameter. Namely:

$$R = \frac{2\pi rl}{\pi r^2 l} = \frac{4}{d} \tag{1}$$

where, R—contace rate; d—pipe diameter; r—pipe radius; and l—pipe length.

It can be concluded from Formula (1) that contact rate increases with the decrease of pipe diameter, leading to faster chlorine decay rate and more chlorine consumption.

As shown in Fig. 9, for DN50 and DN100, THM increases with the increase of diameter. This is because the bigger the diameter is, the smaller the contact rate is. Thus, chlorine consumption on pipe wall is smaller. So that more chlorine can react with organics in water, and THMs' level is higher.

4. Conclusions

(1) Under the condition of the same initial chlorine, the ranking of THMs concentration in different pipe materials was PVC > stainless steel > ductile iron.

(2) Under the condition of the same diameter and retention time, the variation of THM amount in distribution system simulator is consistent with that of bulk water experiment. THM amount is only slightly higher in distribution system simulator, not as distinct as chlorine decay. THMs amount increases with the increase of flow velocity, but it is not as distinct as chlorine decay.
(3) Under the condition of the same distance and different flow velocities and diameters, when both retention time and velocity influence THM formation, its variation is decided by the influential degree of the two influences.
(4) Under the condition of the same distance and different flow velocities, THM increases with the increase of diameter.
(5) Chlorine residual, pipe materials, flow velocity and retention time are all the influences important for THM formation, but not unique conditions. The formation of THMs in real networks is dependent on various influences.

Acknowledgments

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