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Effects of advanced treatments using granular activated carbon adsorption with ozonation and ultrafiltration on chlorine decay

Younghyo Kim[a], Jihyang Kweon[a,*], Dooil Kim[b], Changha Lee[c]

[a]*Department of Environmental Engineering, Konkuk University, #1 Hwayang-dong, Gwangjin-gu, Seoul, South Korea*
Email: jhkweon@konkuk.ac.kr
[b]*Department of Civil and Environmental Engineering, Dankook University, Jukjeon-dong, Suji-gu, Yongin-si, Gyeonggi-do 448-701, South Korea*
[c]*School of Urban and Environmental Engineering, Ulsan National Institute of Science and Technology, UNIST-gil 50, Ulsan 689-798, South Korea*

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ABSTRACT

The application of advanced treatment processes has been substantially increased to comply with regulations on microbial inactivation and disinfection by-products. The advanced processes, such as ozonation followed by granular activated carbon or ultrafiltration, yield changes in chemical properties of the treated water in addition to the improvement of water quality. The changes in water chemistry could affect the kinetics of disinfectant decay within the water distribution system. In addition, decay behaviors using various pipe materials were investigated with water that underwent advanced treatments. The permeate from ultrafiltration generally shows lower decay rate constants than that of effluents from ozonation + granular activated carbon adsorption. The differences were especially obvious for so-called unreactive pipe coupons such as polyvinyl chloride (PVC), polyethylene (PE), polycarbonate (PC), and stainless steel. Reactive pipe materials, such as cast iron and copper, had almost 10 times higher rate constants than the unreactive pipes, regardless of the applied treatment processes. Appropriate safety actions should be introduced to ensure high quality of drinking water in a distribution system prior to changing processes in water treatment plants.

Keywords: Chlorine decay; Distribution system; GAC and ozone process; Ultrafiltration; Wall materials

1. Introduction

Effective disinfection in a water distribution system is necessary to maintain the safety of drinking water. Chlorine is the most widely used water disinfectant globally due to its relatively low cost, efficient disinfection, and ability to provide residual chlorine concentration in a distribution network. Chlorine, as a non-selective oxidant, can react with both organic and inorganic chemical species in water and protect from infection by water-borne diseases.

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*Corresponding author.

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However, there are increasing concerns about disinfection by-products (DBPs) of chlorination and their harmful effects on human health [1,2]. Therefore, it is important to maintain appropriate chlorine concentration to balance potential risks from formation of DBPs against those of microbial infection [3].

Water quality in a distribution system is determined by various factors, including influent water quality from water treatment plants, the properties of distribution pipes, such as age and raw material, and operational conditions such as temperature, pH, and alkalinity [4]. Water quality within a distribution system is also closely related to the extent of chlorine degradation, which occurs as chlorine moves through distribution pipes. Chlorine decay is directly influenced by hydraulic, chemical, microbiological, and infrastructural factors.

Chlorine decay occurs over time due to reactions with compounds contained within the bulk water (termed bulk decay), or due to reactions at the pipe wall (wall decay). Hallam et al. showed that pipe materials strongly influenced wall decay during distribution, and that pipe materials could be classified as reactive (e.g. unlined iron) or unreactive (e.g. polyvinyl chloride (PVC), medium-density polyethylene (MDPE), and cement-lined iron) [5]. The same study also reported that laboratory experiments to measure decay gave results representative of the *in situ* distribution system. Rossman investigated overall chlorine decay with unlined ductile iron pipe using a distribution system simulator. Overall chlorine decay was consistently dominated by wall reactions, which showed first-order rate constants that were an order of magnitude greater than those for the bulk water [6]. The results suggested that pipe wall materials should be evaluated to understand overall degradation of chlorine in the distribution system. Categorizing chlorine decay as bulk and wall decay simplifies the calibration of a chlorine decay model within software simulations of flows in complex pipe and reservoir networks [7]. A suitable general model of chlorine decay in the transported bulk water is an essential component for efficiently modeling chlorine concentration in distribution systems.

Many studies have attempted to develop an accurate network model of water quality using chlorine decay model, and a number of chlorine decay models have been proposed during the past several decades [2]. The most commonly applied so far, the chlorine bulk decay model, is a pseudo-first-order model. The first-order model is based on an assumption that chlorine reacts at a constant rate with excess chlorine-demanding reactants [1]. Several researchers argued that some of the behavior observed for the chlorine decay contradicts the assumption of first-order kinetics [2,7–9]. Based on extensive literature reviews by Kohpaei et al., a two-constituent decay model was shown in practice, to provide sufficient accuracy, simplicity, and the ability to predict disinfection behavior of the system [2]. To adequately represent chlorine decay for a range of initial concentrations used in real distribution systems, an empirical relationship between rate constant and initial chlorine concentration is required for the water of interest.

The rate of bulk reactions of chlorine in the distribution system is significantly affected by the quality of treated water. There has been substantial recent development of advanced treatment process to comply with microbial inactivation and DBP regulations. Ozone oxidation followed by granular activated carbon adsorption (ozone + GAC) and membrane filtration techniques such as microfiltration and ultrafiltration, have been extensively introduced to water treatment plants. Membrane filtration is a newly applied process to the water industry in Korea. Changes in the quality of treated water (influent water to a distribution system) could have effects on the kinetics of disinfectant decay in the distribution system. Compared to conventional treatment process, advanced treatment processes include several operation differences that can yield substantial changes in water quality, such as low dose of coagulant, high final pH, and concentrations of natural organic matter. Rossman investigated the effects of advance treatments, including GAC adsorption, ozonation, and reverse osmosis process, on metallic pipes. It was concluded that the wall rate constants for ozonated and GAC-treated water were approximately twice those of conventional or reverse osmosis-treated water. However, the study was limited to metallic pipes [6]. The objectives of this study are to evaluate the effects of advanced treatment processes, i.e. ultrafiltration, and ozonation followed by GAC, on chlorine degradation; and to examine the effects of wall materials on overall chlorine decay. The first-order decay model (FDM) and the two-constitute decay model (TCM) were compared for properties of chlorine decay between advanced and conventionally treated waters using different pipe wall materials. The results are applicable to chlorine decay behaviors in a distribution system when the treatment process is changed.

2. Methods and materials

2.1. Water samples

Three water samples were tested as influent to a distribution system. Distilled water from a deionizer (ELGA Labwater, PURELAB) was used as a control containing no organic or inorganic matter. Effluent from an ozonation + GAC adsorption process was taken from a pilot plant in Ansan water treatment plant, which intakes water from Paldang Dam and applies conventional treatment processes including coagulation and filtration. The permeate from ultrafiltration was produced by a stirred batch cell reactor in the laboratory. The batch reactor was a dead-end cell reactor (Millipore Co., USA). Transmembrane pressure was held at 30 psi and 3.0 L of permeate volume was produced. The ultrafiltration disc filter (Ultracel, Amicon) was made from regenerated cellulose material, and has a nominal molecular weight cutoff of 30 kDa. Key qualities of the waters used in this study are summarized in Table 1.

2.2. Experimental procedure

Each experiment began by preparation of pipe coupons and a NaOCl stock solution. Six coupons (Biosurface, USA) were washed three times with deionized water and dried in oven for 30 min at 121 ℃. The prepared coupons were stored in a desiccator until use. Each coupon has a diameter of 12.7 mm, a thickness of 3 mm, and a weight of 0.44–4.25 g depending on raw materials. The coupons were made of cast iron (CI), copper (CU), polyethylene (PE), polyvinylchloride (PVC), polycarbonate (PC), or stainless steel 304 (SS). Chlorine dosage of 10 mg/L was used to represent a scenario with poor distribution conditions such as long distance and duration. This concentration was relatively high compared to the normal 4–6 mg/L dose in water treatment plants; however, it is within the range of concentrations encountered in practice. One coupon was added to each 45 mL polypropylene (PP) tube of treated water. The tubes with coupons were stored in darkness at 20 ℃. The waters were taken from the tubes approximately once daily to measure water quality. The experimental procedure is shown in Fig. 1. Residual chlorine was measured by DPD colorimetric method followed by standard method [10]. The standard curve was obtained with 0, 2, 4, 6, 8, and 10 mg/L of free chlorine. Some samples with high turbidity were prefiltered using a 0.45 μm polypropylene syringe filter (Millipore Co., USA). Water parameters were measured via a pH meter (Thermo 3 star, USA), conductometers (YSI-3100 and Thermo 5 star, USA), TOC analyzer (SIEVERS 5310C, USA), turbidimeter (Hach 2100N, USA), and UV/VIS spectrophotometer (Thermo GENESYS 10UV, USA).

Table 1
Quality parameters of intake water for UF, UF permeate, and ozone + GAC-treated water

	Intake water	UF permeate	Ozone GAC-treated
Temperature (℃)	28	27	27
pH	8.33	8.60	8.30
Conductivity (μs/cm)	183.8	177.8	186.1
Turbidity (NTU)	7.52	0.298	0.187
Alkalinity (mg $CaCO_3$/L)	36.4	39.0	32.8
DOC (mg/L)	2.38	2.04	1.05

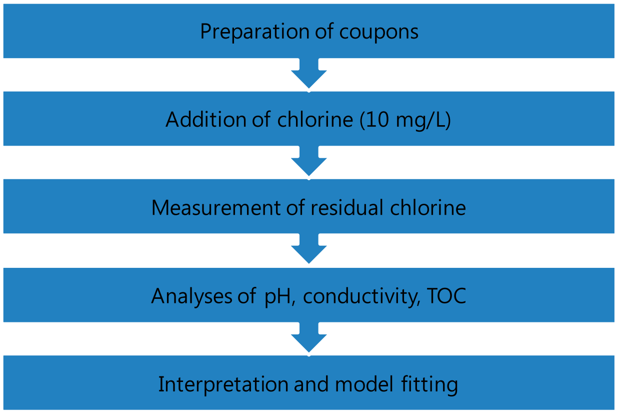

Fig. 1. Experimental procedure for chlorine decay experiments.

2.3. Model description

The first-order chlorine decay model (FDM) can be defined as shown in Eq. (1). The general analytical solution for the first-order kinetic model to predict the decrease in concentration of chlorine in water is expressed as shown in Eq. (2).

$$\frac{dC_{\mathrm{Cl}}}{dt} = -k \times C_{\mathrm{Cl}} \tag{1}$$

$$C_{\mathrm{Cl}}(t) = C_{\mathrm{Cl},0} \times \exp(-k \times t) \tag{2}$$

In the equation, $C_{\mathrm{Cl}}(t)$ is the chlorine concentration at time t (mg/L), $C_{\mathrm{Cl},0}$ is the initial chlorine concentration at time $t = 0$, and k is the first-order decay constant (h^{-1}). The first-order reaction rate constant was obtained with the least-square regression method.

TCMs were developed by several researchers. The model proposed by Clark was simplified by Kohpaei et al. to provide an analytical solution [2,11]. The simplified reaction and the solution are shown in Eqs. (3) and (4):

$$\mathrm{Cl}_2 + RA \xrightarrow{k} \text{inert product} \tag{3}$$

$$C_{\mathrm{Cl}}(t) = \frac{C_{\mathrm{Cl}_0} - C_{\mathrm{RA}_0}}{1 - (C_{\mathrm{RA}_0}/C_{\mathrm{Cl}_0}) \times \exp(-(C_{\mathrm{Cl}_0} - C_{\mathrm{RA}_0}) \times k_2 \times t} \tag{4}$$

where RA represents all agents that react with chlorine in the distribution system and $C_{\mathrm{RA},0}$ is the initial concentrations of the hypothetical reagents. The second-order reaction rate constant (k_2) was calculated using a solver program in MS Excel software.

The sum of the squares of the residuals or errors (SSE) between the data points and prediction was computed followed by Eq. (5). The SSE represents the uncertainty that remains after the fitting experimental results with model prediction. Also, the weighted error between the experimental and model data (χ^2) was used as one of the measures of the goodness of fit between experimental and predicted data as shown in Eq. (6).

$$\begin{aligned} \mathrm{SSE} = S_{\mathrm{r}} &= \sum_{i=1}^{n} e_i^2 = \sum_{i=1}^{n} (y_{i,\,\mathrm{measured}} - y_{i,\,\mathrm{model}}) \\ &= \sum_{i=1}^{n} (\exp - fit_i)^2 \end{aligned} \tag{5}$$

$$\chi^2(p) = \sum_{i=1}^{n} \left(\frac{y_{i,\,\mathrm{measured}} - y_{i,\,\mathrm{model}}}{\sigma_{\mathrm{measured},i}} \right)^2 \tag{6}$$

where $y_{i,\mathrm{measured}}$ is the ith measured value, $y_{i,\mathrm{model}}$ is the calculated value form the model, and $\sigma_{i,\mathrm{measured}}$ is the standard deviation of $y_{i,\mathrm{measured}}$.

3. Results and discussion

3.1. *Effects of treatment characteristics on chlorine bulk decay*

The distilled, UF-treated, and ozone + GAC treated waters were introduced to the blank tubes and residual chlorine was analyzed as shown in Fig. 2. The tube was used to ensure that chlorine reacted with substances in the bulk water, avoiding reaction with pipe wall materials. The data were then fitted to the FDM and the TCM. During experiments with the distilled water, infinitesimal amounts of chlorine were degraded indicating negligible concentration of reactive substances in the bulk. The other two waters showed some extent of chlorine degradation. Chlorine decay by the UF permeate occurred gradually throughout the entire duration of the experiments, whereas the decay by the ozone + GAC effluent was seemingly completed during the first 24 h. The difference might be due to the amounts and properties of substances to be reacted with chlorine. The DOC concentration was higher with the UF permeates than ozone + GAC effluent as shown in Table 1. In addition, ozone oxidation has been known to decompose high-molecular weight organics to low-molecular weight organics, which could result in greater probability of reaction with chlorine than in the permeate from ultrafiltration. As the main removal mechanism in membrane filtration is physical sieving through the pores in the filter, there is limit to change the chemical properties of UF permeate during this process. In addition, the two-constituent decay models were shown to better fit the experimental data than the FDM. The goodness of the fit was noticeable with ozone GAC effluent whereas the distilled water showed little difference with two model predictions.

The rate constants from the models are summarized in Table 2. The k_1 values from the UF permeate and ozone + GAC effluent were similar compared to the small k_1 value of the distilled water. The greater k_2 value of the ozone + GAC effluent than the UF permeate was consistent with the graphical expression in Fig. 2. The weighted errors, χ^2, showed a smaller value (1.4988) in the two-constituent decay model compared to that of the FDM (18.656). The results indicated that the two-constituent decay model provided a more reliable representation of chlorine decay phenomena than the FDM. As described in the model

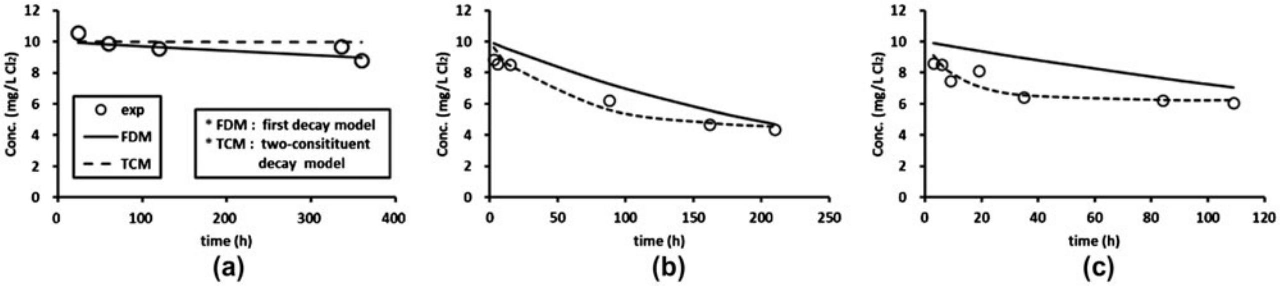

Fig. 2. Chlorine bulk decay with (a) Distilled water, (b) UF permeate, and (c) Ozone + GAC effluent.

Table 2
Model parameters from tested waters in glass tubes

Model parameters	Water samples		
	Distilled water	UF permeate	Ozone GAC effluent
k_1 (h^{-1})	0.0003	0.0036	0.0032
k_2 ($(mg/L)^{-1}(h^{-1})$)	0.0000	0.0022	0.0093
χ^2 (k_1)	1.0200	0.2826	18.656
χ^2 (k_2)	0.6934	0.2734	1.4988

section, the model accounts for chlorine reactions with any compound in the bulk. The hypothetical reactions considered in the model do not represent the actual mechanism. The approach in the model is expected to be consistent with conceptual chorine decay kinetics. However, this involves two known drawbacks: one is that the proposed analytical solution is only valid when the initial chlorine concentration is not the same as that of the reactive substances (which are designated as $C_{RA,0}$); secondly, it considers only one individual species to react with chlorine. Despite these limitations, the two-constituent decay model is still promising due to its simplicity in hypothesis, and the analytical solution is available [7].

3.2. Chlorine decay behaviors for different pipe materials

The chlorine decay patterns with six pipe wall materials were evaluated using distilled water as shown in Fig. 3. The degree of chlorine decay differed substantially depending on wall materials. Chlorine concentrations decreased rapidly when CI and CU coupons were added to the tubes. In contrast, plastic materials including PE, PVC, and PC, and SS coupons showed relatively gradual decrease of chlorine concentrations. According to Hallem et al., the pipes used in this study could be classified as reactive (CI and CU) and likely unreactive (PVC, PE, PC, and SS) [5]. The work by Hallem et al. also concluded that the decay of chlorine in unreactive pipes was limited by the reactivity of the pipe material, whereas chlorine decay in reactive pipes was limited by the transfer of chlorine to the pipe wall. In the present study, relatively high concentrations of chlorine were used, which yielded considerable transfer of chlorine to the pipe wall and significant chlorine decay with reactive pipes, i.e. cast iron and copper coupons.

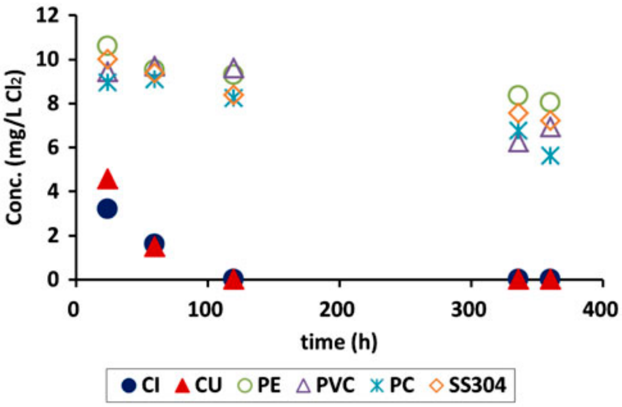

Fig. 3. Chlorine decay by various pipe wall materials tested in distilled water.

Zhang et al. investigated the effect of copper corrosion products on chlorine degradation and haloacetic acid (HAA) formation [12]. They found that chlorine decay and HAA formation were significantly enhanced in the presence of copper. The extent of copper catalysis was greatly affected by the pH of the solution and the concentration of copper corrosion products. Accelerated chlorine decay was observed at pH 8.6 compared with pH 6.6 and pH 7.6. The pH used in this study was usually maintained at relatively alkaline conditions (as shown in Table 1), which might induce the rapid decay of chlorine in the presence of copper coupons.

The kinetic results with the tested waters for various wall materials are presented in Table 3. The first-order rate constants with distilled water clearly explained the reactivity of the wall materials. The value was 0.0311–0.019 h^{-1} for reactive pipes and 0.0007–0.0013 h^{-1} for unreactive pipes. In the previous study, when the catalyzed corrosion by copper occurred, the first-order reaction rate, k_1 values, was reported within the range of 0.012–0.584 h^{-1} [12]. The results of rate constants from the two-constituent model also corresponded to the decay trend in Fig. 3, although the magnitude was small thus difficult to compare precisely.

3.3. Effects of advanced treatment on chlorine decay in various pipe materials

The most frequently applied advanced treatment processes in Korea is ozonation followed by granular-activated carbon. Several consumers complained about water quality after water treated by advanced processes was supplied. Taylor et al. investigated the effects of changes of source water on the quality of distribution water, especially on metal release by corrosion [13]. They reported that aged cast iron pipes that carried conventionally treated ground water released significantly greater amounts of iron when exposed to desalinated seawater or surface water processed by enhanced treatment. It was suggested that

Table 3
Kinetic results for distilled waters with six different pipe wall materials

Parameters	PVC	PE	PC	SS	CU	CI
Distilled water						
k_1 (h^{-1})	0.0013	0.0007	0.0013	0.0008	0.0311	0.019
k_2 $(mg/L)^{-1}(h^{-1})$	0.0000	0.0000	0.0002	0.0004	0.0002	0.0052
χ^2 (k_1)	0.8426	1.0200	0.6743	0.5781	0.0294	8.2518
χ^2 (k_2)	0.6382	0.6935	0.5038	0.2110	0.0146	0.1759

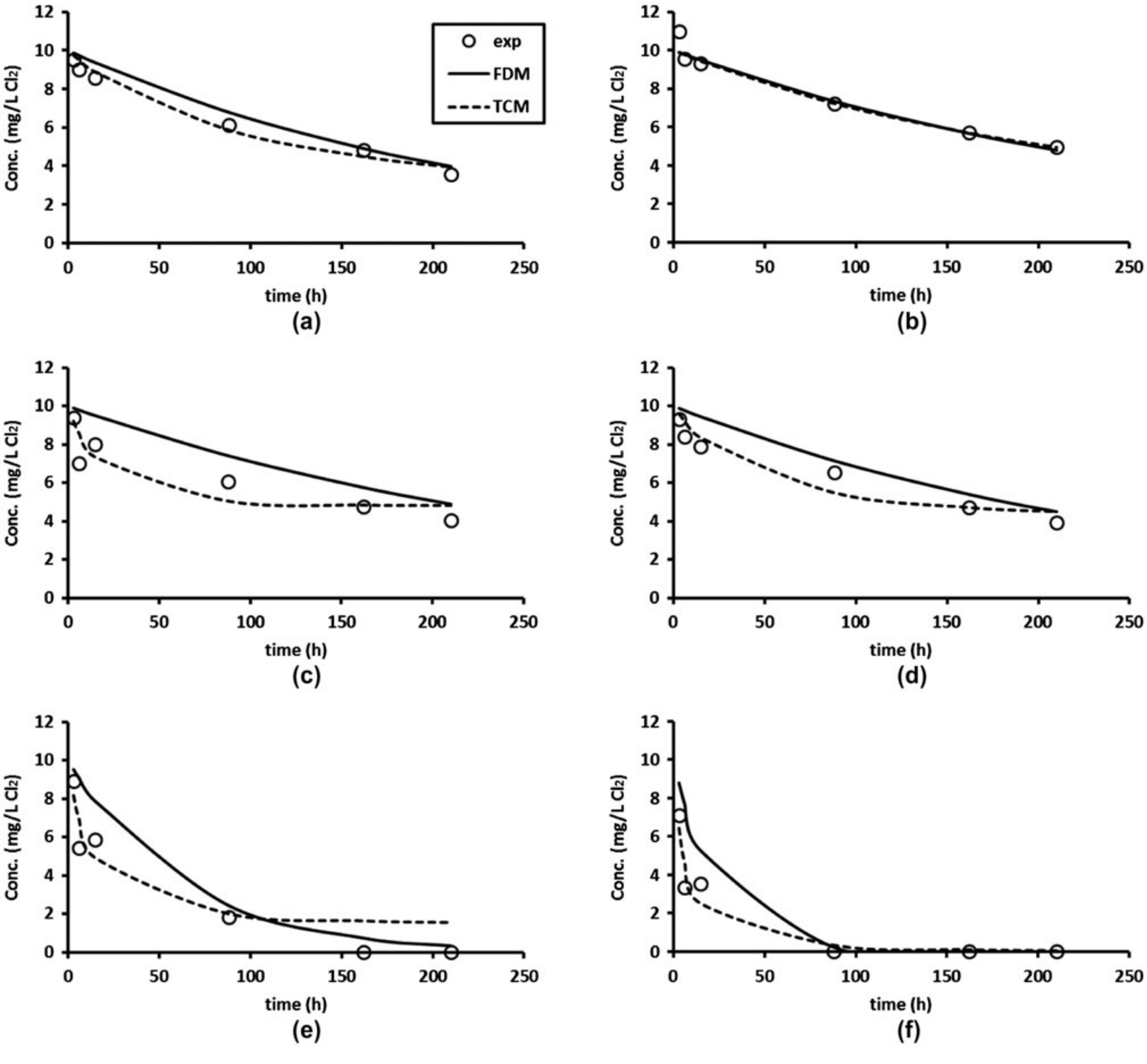

Fig. 4. Chlorine decay by UF permeate with various pipe wall materials (a) PVC, (b) PE, (c) PC, (d) SS, (e) CU, and (f) CI.

increased iron release was initiated by alkalinity levels lower than that of the ground water. Parameters such as pH, alkalinity, dissolved oxygen content, and ion concentrations of chloride and sulfate were reported to influence the quality of distribution water greatly [13,14].

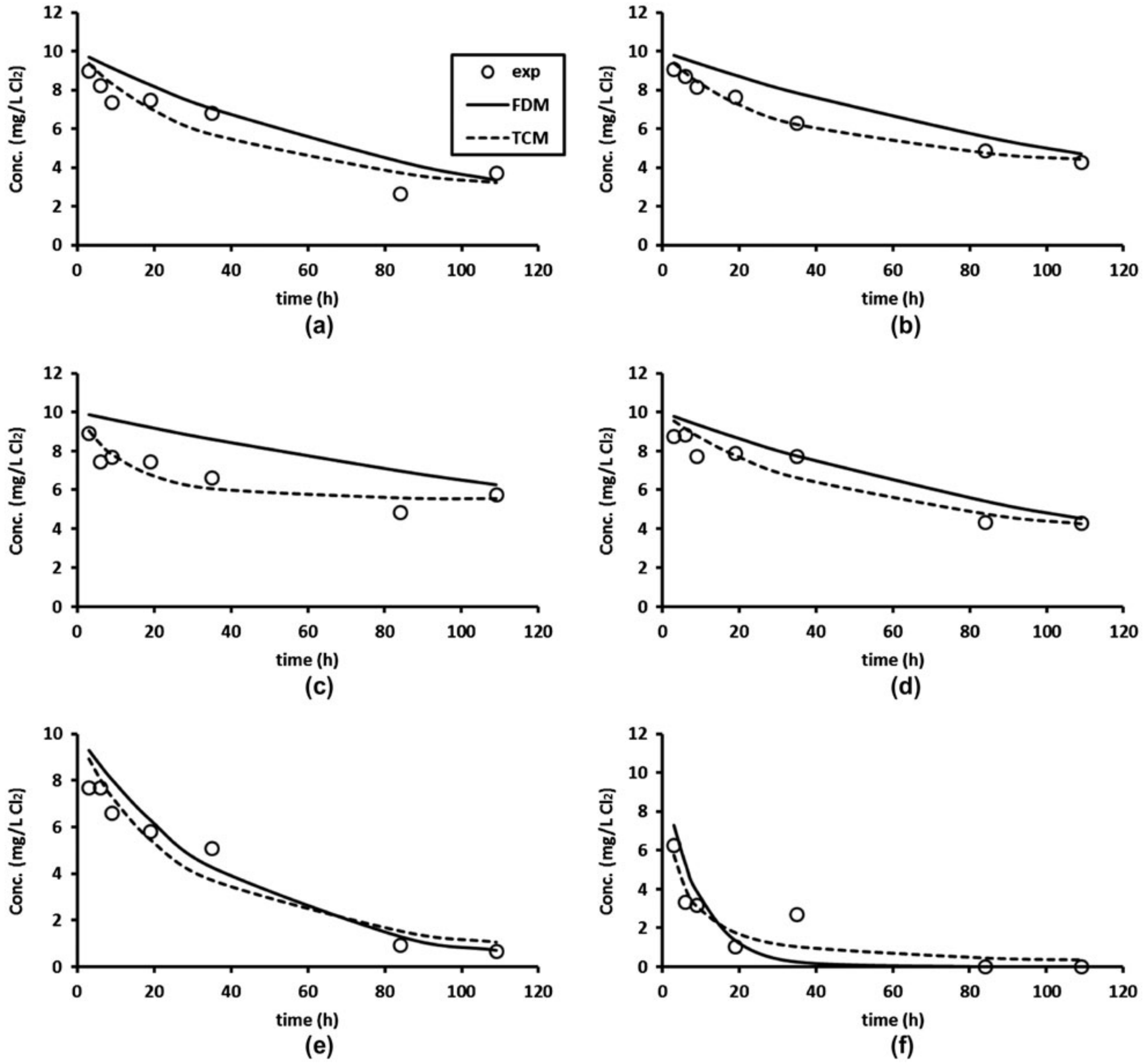

Fig. 5. Chlorine decay by ozone + GAC effluent with various pipe wall materials (a) PVC, (b) PE, (c) PC, (d) SS, (e) CU, and (f) CI.

The UF permeate and ozone + GAC effluents were added to the experimental tubes with pipe coupons of different wall materials. The chlorine decay results are shown in Fig. 4 for the UF permeate and Fig. 5 for the ozone + GAC effluent. Two models were used to find the best fit for the rate constants of chlorine decay, which are summarized in Table 4.

Fig. 4 shows that so-called reactive pipe materials reacted greatly with chlorine, which thus showed greater decline than in the unreactive pipes, as expected. The degree of decay with the UF permeate was likely greater than that with distilled water, as also indicated by the rate constant values of the two kinetic models. The first-order chlorine decay rates of the unreactive pipes were within the range of 0.0034–0.0044 h^{-1}, which were lower than the range of 0.016–0.043 h^{-1} for the reactive pipes. The k_1 values of the UF permeate were generally greater than those for distilled water. The k_2 values were more recognizable with the UF permeate and ranged in 0.0002–0.0169 $(mg/L)^{-1}(h^{-1})$ compared to the distilled water, which spanned in the range of

Table 4
Kinetic results for the advance treated waters with six different pipe wall materials

Parameters	PVC	PE	PC	SS	CU	CI
UF permeate						
k_1 (h^{-1})	0.0044	0.0035	0.0034	0.0038	0.016	0.043
k_2 $(mg/L)^{-1}(h^{-1})$	0.0010	0.0002	0.0057	0.0025	0.0089	0.0169
χ^2 (k_1)	0.3763	0.2826	4.0987	1.5254	2.9781	3.5980
χ^2 (k_2)	0.1472	0.2734	1.3936	0.6492	1.4359	0.4963
Ozone GAC effluent						
k_1 (h^{-1})	0.0100	0.0069	0.0043	0.0073	0.0243	0.1053
k_2 $(mg/L)^{-1}(h^{-1})$	0.0026	0.0035	0.0085	0.0020	0.0028	0.0246
χ^2 (k_1)	1.7733	2.3074	13.850	1.8788	0.8563	2.6782
χ^2 (k_2)	0.8750	0.1147	1.4041	0.9909	0.6053	1.0230

0–0.0052 $(mg/L)^{-1}(h^{-1})$. According to Kohpaei et al., the values of the distilled water corresponded to the rate constants for slow reducing agents in the water [2]. In addition, the experimental data were more closely fitted to the two-constituent decay model than the first-order model, indicating the smaller the weighted error between the experimental and the TCM data in most cases.

The chlorine decay results in Fig. 5 showed that the most rapid degradation occurred for the water treated by ozone + GAC adsorption. The *x*-axis represents experiment duration and reached 120 h, which is shorter than that for other waters. For example, in the case of PVC pipes, approximately 80 h were required to obtain 3 mg/L of residual chlorine for the ozone + GAC effluent, whereas more than 200 h were needed for the UF permeate. A study by Rossman on the effects of advanced treatment on chorine degradation reported that the type of treatment had clear effects on the bulk water reaction of free chlorine, showing changes in the reaction rate constant [6]. The rate constant followed the sequence: reverse osmosis treatment, > GAC, > ozonation, > conventional treatment and the latter two treatments were almost equivalent. For the wall reaction, the rate constants for conventional and RO-treated waters were 0.0442–0.1146 h^{-1} and those for ozone + GAC-treated water were 0.123–0.192 h^{-1}. The total chlorine reaction rate was highest with ozonated water at ozone doses of 4 mg/L. In this study, the rate constants of FDM for the reactive pipes were 0.0243–0.1053 h^{-1} and those for the unreactive pipes were 0.0043–0.01 h^{-1}, which showed almost doubling of the rate constants for ozonation + GAC-treated water compared to the UF permeate. Again, the two-constituent decay model fitted better with the experimental data. It should be noted that chlorine decay could not be explained by a pure first-order process, but was clearly influenced by the concentrations of the species reacting with chlorine.

4. Conclusions

The effects of the advanced treatments on chlorine decay with various pipe wall materials were investigated to understand chlorine reaction in the bulk water and wall surfaces in distribution systems. The bulk decay rates by the first-order kinetics of waters from the advanced processes (ultrafiltration; ozonation followed by GAC) were within the range of 0.0032–0.0036 h^{-1}. The total decay rates of FDM for the waters varied greatly with different pipe materials, which were in the range of 0.0034–0.1053 h^{-1}. Both models showed similar reactivity trend for the effects of tested waters on bulk (using PP tube) and on total (using various) pipe materials. Compared to the first-order decay kinetics, the two-constituent decay models showed better reproduction of the experimental results in most cases. The two-constituent decay model should be incorporated into existing distribution models. It was also clear that the ozone + GAC effluent generally had higher rate constants than the UF permeate. Great caution is required when ozonation followed by GAC is applied to distribution systems because the great reactivity might make a place in the distribution system in which a chlorine concentration is insufficient to satisfy inactivation of microbial regrowth. The results will improve water disinfection operations and also be useful in maintaining residual chlorine levels throughout distribution systems.

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