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Free Chlorine Concentration Decay Rate with Fast and Slow Reacting Agents Along with Service Age of the Distribution System

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Abstract

One of the most crucial elements influencing the rate of chlorine decay in drinking water networks is the service age of pipes of the distribution system, which has been introduced in this study. The aim of the study was to demonstrate the effect of the distribution network's service age and the chlorine degradation rates with the concentration of slow- and fast-reacting agents which were investigated under the fixed temperature of 30 degrees centigrade and the results are displayed with the help of various graphs. It was found that with ageing factor of 0.01 chlorine concentration took 16 hours to become zero while on the other hand with ageing factor of 0.4 the chlorine concentration reduced to zero in 8 hours. Through correlation analysis a very strong positive correlation was found to be 0.9830450 with ageing factor of 0.1 for newly built pipeline on the other hand it was found to be slightly lower with the value 0.978139 with ageing factor of 0.4 for old pipeline.

Keywords- Arrhenius equation, Kinetic modelling of chlorine decomposition, Service age coefficients and reactant agents.

Nomenclature

Parameters		**Unit**
C_{cl}	The amount of free chlorine	mg/l
C_f	The amount of fast-reducing agents	mg/l
C_s	The amount of slow-reducing agents	mg/l
k_f	The amounts of fast response rates	$l/(mg.h)$
k_s	The amounts of slow response rates	$l/(mg.h)$
k	Ageing factor	h
k_T	The reaction rate coefficients at a specific temperature	$l/(mg.h)$
E_A	Activation energy	KJ/mol
R	Universal gas coefficients	$J/kelvin.mol$
T	Temperature	℃

1. Introduction

In many water distribution systems, a major distribution goal is to maintain chlorine residual by effectively modeling chlorine concentration. The secondary disinfection of drinking water distribution system aims at maintaining chlorine residuals from the entry point to the farthest point of the system (USEPA, 2002; WHO, 2004). The chlorine is lost by the reaction with organic matter and inorganic substances like ammonia, manganese, or iron left in the water after treatment, hence a higher concentration of chlorine is required at the entry point as compared to the extremities. With the higher entry concentration of chlorine, the odour and change in the taste may be generated at upstream locations that need elimination through downstream booster dosing or lower chlorine concentration at extremities (USEPA, 2006; WHO, 2004). In the planning and management of system hydraulic performance nowadays software packages are used to simulate flows in complex pipes or reservoir networks. The chlorine decay in pipes can be divided into two parts, bulk decay and wall decay. The bulk decay results from the reaction of chlorine substances in the bulk water. On the other hand, wall decay occurs due to the reaction of chlorine with the pipe wall and the biofilm as well as corrosion products that stick to them. The disinfectant residual used most frequently to maintain the microbiological safety of drinking water in supply networks is chlorine. Chlorine concentration must be maintained within a very small range, typically between 0.2 and 1.0 mg/l, to be effective and to prevent the overproduction of toxic disinfection by-products (WHO, 2011). Because chlorine concentration decreases as water moves through transport and distribution networks, managing chlorine residuals is not an easy task. For the maintenance of water quality in transmission and distribution networks, disinfecting drinking water is essential. Before entering the transportation system, treated water is cleaned and disinfected. A detectable disinfectant residual should stay in the water because bacterial contamination of water is anticipated in the transmission and distribution system, reducing the risk of waterborne illness and biofilm development (Clark and Coyle, 1990). Water Research Centre (1976) had previously recommended some factors to take into account for controlling the number of bacteria in treatment and distribution systems, including system cleaning, limiting the amount of time water is retained in the network, and using alternative disinfectants that may last longer in the water. According to Kiene et al. (1998), this degradation is primarily caused by chemical reactions between chlorine and natural organic matter (NOM) dissolved in the water (bulk decay) and internal surface materials of the pipes (wall decay). The difficulty of predicting the disinfectant's concentration in supply systems water through modeling is increased by the fact that decay rates are influenced by a variety of factors, including temperature, initial chlorine concentration, NOM nature and concentration, as well as hydrodynamic conditions (Powell et al., 2000). Menaia et al. (2003) stated that chlorine decay in the pipe depends on the water flow velocity and the pipe radius. Many more hydraulic conditions are dependent on chlorine decay in water. Brown et al. (2011) studied that the presence of free chlorine is the prior condition for trihalomethanes (*THMs*). Chlorine residual modeling is extremely helpful for controlling disinfectant concentrations in transportation and distribution networks, but it heavily depends on how accurately chlorine decay is described. Fisher et al. (2011a) found that the first reactant model is non-suitable for analyzing the chlorine decay in the water distribution network as compare to the second reactant model. Temperature has been identified as one of the most important variables affecting chlorine decomposition rates and should be included in decay models (Fisher et al., 2011b). Jadas-Hécart et al. (1992) used the SR model to decay tests conducted at different temperatures on two samples of treated water, taken at an interval of a year and acknowledged that in simple reactions, the decay coefficient increases with temperature in accordance to the Arrhenius formula. However, the SR model did not meet the invariance of parameters criteria with respect to either time or temperature. Fisher and Kastl (1996) and later Kastl et al. (2003) used an augmented four reactant (*4RA*) model to study the effect of temperature on second order decay coefficient for water from different locations and cited corresponding *E/R* values. In the context of ongoing climate change, especially in the northern latitudes, increases in average water temperature in the supply systems are predicted, in addition to significant seasonal variations (Sérodes et al., 2003; Uyak & Toroz, 2006).

The Arrhenius gave a mathematical expression, which is used to describe the rate of a chemical reaction under the given temperature. In this expression k is the reaction rate, A is the frequency of molecules, E is known as the activation energy for the reaction, R is considered as the ideal gas constant and T is the absolute temperature.

Kinetic modeling of chlorine decomposition in the distribution system involves the following situations.

(i) In the first order kinetics the first order reaction is involved and chlorine concentration is found to be directly propositional to the rate of chlorine decomposition.
(ii) In the second order kinetics decay the chlorine concentration decay is proportional to its concentration and other factors that are involved in the reaction.
(iii) The temperature dependence in the kinetic modelling is used with the help of Arrhenius equation.

It is noticed that according to service age coefficients, the chlorine level of water utilities remains the same within the effective ranges.

Constant for chlorine wall disintegration qualifies the rate at which chlorine concentration is decreased by the reaction with the wall of the distribution system. It involves pipe age and the pH of the water, pipe material, and flow conditions. The constant for chlorine wall disintegration is denoted by the k_w.

Fast-reactant agents: Fast-reactant agents are used to speed up the chlorine reaction with water. There are many more factors that speed up the reaction rate. Humic and fulvic acids are two natural organic materials, which are present in the water. These compounds react with chlorine and form disinfection by-products. Inorganic compound nitrate is often present in the water and it involves the reaction of chlorine with water. Biofilm also plays an important role in protecting the water from disinfection by the embedded pathogens.

Slow reactant agents: Slow-reactant agents are used to control the chemical reaction that ensure to supply the of pure and quality water to every human. Cyanuric acid is used as slow reactant agent in swimming pools, which reduces the degradation of chlorine in the water.

The observational Arrhenius equation describes the impact of temperature on chemical reaction rate coefficients $k = Ae^{-(E_A/RT)}$, where, k is the reaction rate coefficient, A is the frequency factor, E_A is the activation energy, R is the ideal gas constant [8.31 J/(mol K)], and T is the temperature. Each chemical process has its own unique Arrhenius parameters (A and E_A). According to the transition state theory, E_A is the minimal amount of energy needed for a reaction to take place for activated reactant complexes to form during the reaction. High values of E_A typically indicate greater sensitivity of reaction rates to temperature changes. The Arrhenius equation is used to effectively explain how temperature affects the first-order chlorine bulk decay coefficient (Courtis et al., 2009; Hua et al., 1999; Powell et al., 2000). Kastl et al. (1999) stated that the water's NOM comprises two main types of compounds one that reacts quickly with chlorine (fast reducing agents), one that reacts slowly (slow reducing agents), that can be used to describe this decay profile. The concentration of free chlorine with time is given by

$$\frac{dCcl}{dt} = \frac{dCf}{dt} + \frac{dCs}{dt} = - k_f C_{cl} C_f - k_s C_{cl} C_s \tag{1}$$

where, C_{Cl} is the concentration of free chlorine (mg/l), C_F and C_S are, respectively the concentrations of fast and slow reducing agents (mg/l), and k_f and k_S are the fast and slow reaction rate $l/(mg.h)$, respectively.

Fisher et al. (2012) augmented the *2R* model by incorporating the temperature effect on the reaction rate coefficients. The authors used the Arrhenius equation to determine the reaction rate coefficients at a given temperature (k_T as a function of each k's base value at a reference temperature of 20C (k_{20}).

$$k_T \;= k_{20} e^{\frac{-\frac{EA}{R(T-20)}}{(273-20)(273-T)}} \tag{2}$$

In the *2RA* model, k_{f20}, k_{s20}, and k are the fast, slow reaction rate and service coefficients at the reference temperature of 20ºC. This method enables the rate factors at a reference temperature to replace the *A* parameter of the Arrhenius equation. The other parameters of the model are effectively used in the current study. Kohpaei & Sathasivan (2011) had also addressed this problem and came to the conclusion that when using a parallel second order model to describe chlorine decay, the use of different or single E_A/R for fast and slow reactions led to similar findings. For a variety of untreated and treated waters from various sources, the impact of temperature on chlorine decay rates was examined in the current study. Gaining knowledge of how temperature affects chlorine decay in waters with various NOM types and determining whether rapid, slow, and pipe aging factor decay phases are impacted by temperature in different ways were the goals of the study. Bhadula et al. (2021) presented a mathematical model, in which Arrhenius model was augmented with the unsteady state mass transfer equation including the aging factor of the pipeline. The chlorine profile was analyzed with the help various of velocities of water, temperature, and wall reaction coefficient, and found that chlorine decreases gradually up to temperature 20 °C but at higher temperatures, chlorine decreases rapidly. Maleki et al. (2023) studied statistical approach to study the chlorine behavior in water pipeline network and they identified that, in the older pipe line system, upto 97 % chlorine consumed by the pipe wall while the role of bulk reaction can be reached about 35 % in the new pipe line. Maphanga et al. (2024) studied chlorine demand in the drinking water distribution system. They analyzed that the reaction with the bulk water consumed a large amount of chlorine and the remaining amount of chlorine was consumed with tank surface and wall.

In the paper we have added the ageing factor of the pipe to the fast and slow reactant agents and then studied the three-parameter model for the water distribution system. Further, the model also shows the effect of temperature on the decay coefficient as per the Arrhenius model.

2. Mathematical Modelling

The general equation for chlorine concentration decay in a water pipeline network with three reactant parameters models can be written as

$$\frac{dCcl}{dt} = \frac{dC_f}{dt} + \frac{dC_s}{dt} + \frac{dC_p}{dt} \tag{3}$$

where, $\frac{dC_f}{dt} = \; -k_f\, C_{cl}\, C_f$, $\frac{dC_s}{dt} = -\, k_s\, C_{cl}\, C_s$ and $\frac{dC_p}{dt} = -\, k\, C_{cl}$ (4)

Substituting Equation (4) in Equation (3), now Equation (3), can be written as

$$\frac{dCcl}{dt} = -k_f\, C_{cl}\, C_f - \; k_s\, C_{cl}\, C_s - \; k\, C_{cl} \tag{5}$$

when

$C = C_0$ at $t = 0$ (6)

Here C_{cl} is the amount of free chlorine (mg/l), C_f and C_s are the amounts of fast and slow reducing agents mg/l, respectively, and k_f, k_s and k are the amounts of fast, slow response rates and aging factor as well as the service coefficients, respectively.

Using the Arrhenius Coefficient, Fisher et al. (2012) calculate the reaction rate coefficients at a specific temperature ($k_T = k_{20}\, e^{\frac{-\frac{E_A}{R(T-20)}}{(273-20)(273-T)}}$) as a function of each $k's$ base value at a reference temperature of 20°C (k_{20}). Thus Equation (5), can now be written as

$$\frac{dCcl}{dt} = -k_{f20}\, e^{\frac{-\frac{E_A}{R(T-20)}}{(273+20)(273+T)}} C_{cl}\, C_f - k_{s20}\, e^{\frac{-\frac{E_A}{R(T-20)}}{(273+20)(273+T)}} C_{cl}\, C_s - k\, C_{cl} \tag{7}$$

Solving Equation (7) analytically, we get

$$C_{cl} = C_0\{exp\,(-k_{f20}\, e^{\frac{-\frac{E_A}{R(T-20)}}{(273+20)(273+T)}} C_f - k_{s20}\, e^{\frac{-\frac{E_A}{R(T-20)}}{(273+20)(273+T)}} C_s - k\,)\}t \tag{8}$$

Various graphs have been plotted using the analytical solution of the model in which an additional parameter of ageing of the pipe was incorporated.

3. Geometry of the Model

The samples for chlorine have been collected from households, where relatively smaller size of pipes are used for the water flow. From **Figure 1**, S1, S2 and S3 are the nodes on the pipeline representing the location for water sample collection to check the chlorine concentration decay.

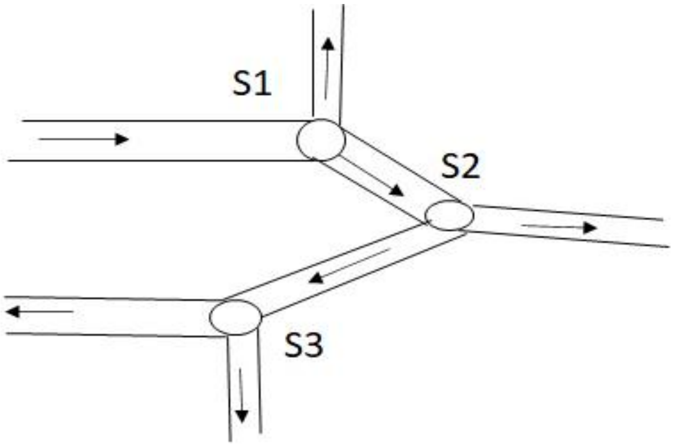

Figure 1. Geometry of the model.

4. Results

The chlorine concentration degradation with respect to time affected by the service rate coefficient have been computed and graphically displayed using SCI lab software.

This model uses the service rate coefficient to compute the rate of change in chlorine concentration degradation over time. It also considers the effects of slow- and fast-reacting agents in chlorine concentration decay over time.

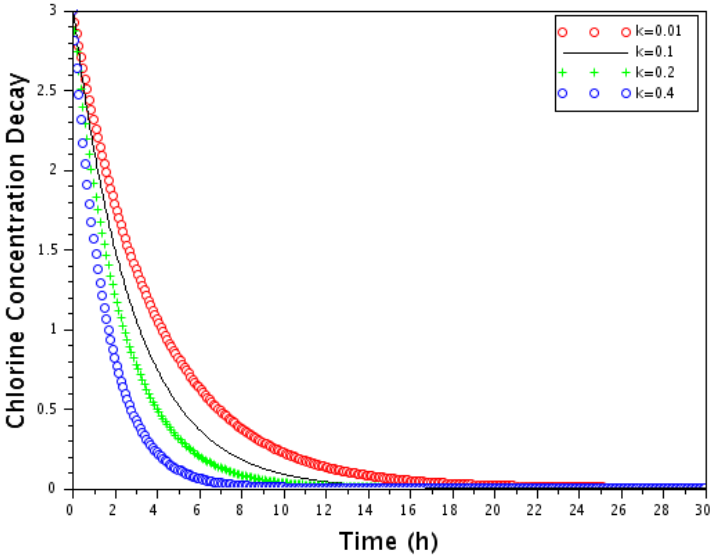

Figure 2. Variation between chlorine concentration decay rate with time for different aging factors is shown.

In **Figure 2**, the chlorine concentration decay is plotted with respect to time, using different values of service age coefficient k (0.01, 0.1, 0.2, and 0.4). The values taken for all service age coefficients $C_f = 0.822$, $C_s = 4.15, k_f = 0.262$ and $k_s = 0.008$ at temperature, 30 degrees centigrade have been taken. It was found that with the increase in service age coefficient, there is a decrease in the time for chlorine concentration decay. For example, with k = 0.01 all chlorine is lost in 16 hours, while with k = 0.4, all chlorine is lost in 8 hours and is not used completely for clearing the water. Maleki et al. (2023) presented the statistical study on the impact of chlorine decay in newly built as well as old pipe line network and they identified that in old pipelines up to 90% of chlorine concentration is consumed by the pipe wall while in newly built pipeline, it consumed up to 30%.

In **Figure 3**, the chlorine concentration decay is plotted with respect to time using different values of the slow reactant agent $k_s(0.1, 0.2, 0.4\ and\ 0.6)$. The values $C_f = 0.822$, $C_s = 4.15$, $k_f = 0.262$ and $k = 0.01$ at temperature, 30 degrees centigrade have been taken. It was found that with $k_s = 0.1$, free chlorine became zero in just 2 hours, thus leaving very little chlorine even to react with the walls of the distribution system. When the of slow reactant agents is increasing from 0.1 to 0.6 chlorine concentration decay are inversely proportional to each other, which due to the slow reacting agents are present in the water it slows the reaction and most of the chlorine reacts with the pipe wall and other inorganic organic compounds which are present in the water. So, one can say that less amount of chlorine purifies the water in this situation.

In **Figure 4**, Various value of fast reacting agents $k_f\,(0.2, 0.4, 0.6, 0.8)$, service age coefficient, $k = 0.01, C_s = 4.15$, and concentration of fast reacting agent $C_f = 0.822$ are taken. The free chlorine is reduced rapidly when the value of the coefficient of the fast reactant agent is increasing. For example, when the value of the fast-reacting agent is 0.2, then free chlorine becomes zero in 8 hours while when the value of fast reactant agent is 0.8, then chlorine becomes zero in 2 hours. Since fast reaction agents accelerate the decay process so chlorine level in the pipeline reduced as the vales of k_f (fast reactant agents) is increasing. Which can be easily seen in the **Figure 4.**

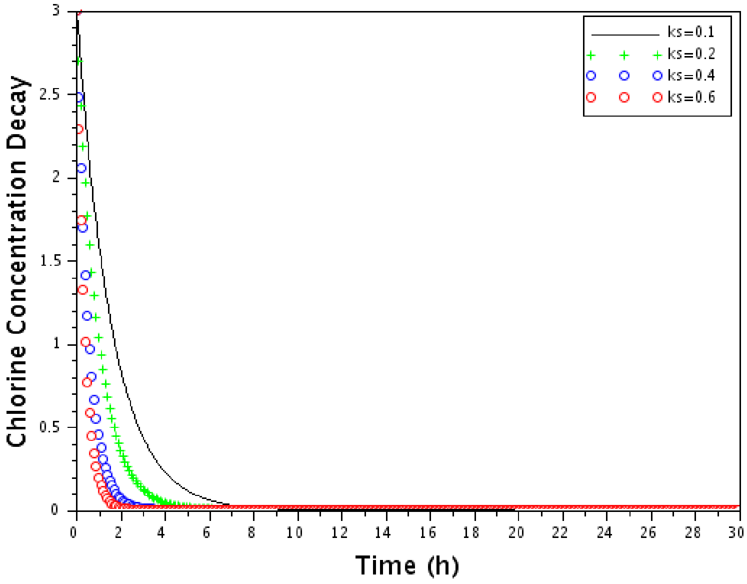

Figure 3. Shows variation between chlorine decay rate with time for different coefficients of slow reactant agents.

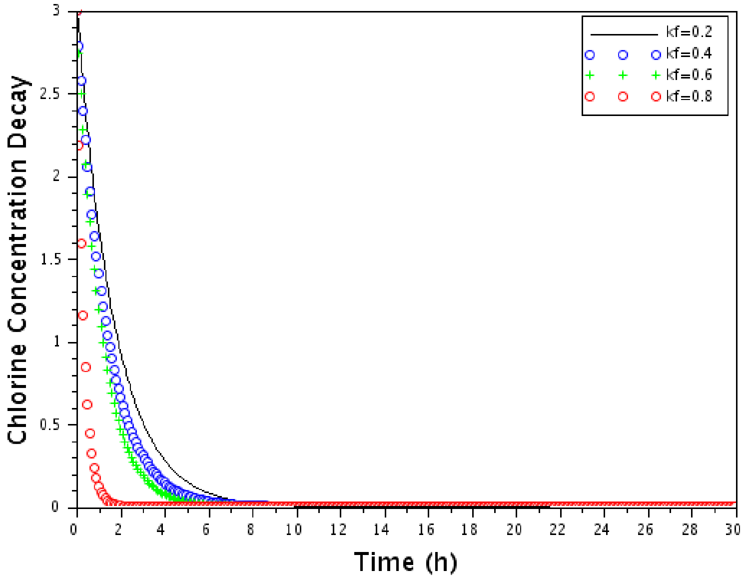

Figure 4. Variation between chlorine decay rate with time for different coefficients of fast reactant agents.

In **Figure 5**, the different values of concentration of slow reactant agent C_s (4.22, 6.0, 8.0 and 10.0), $C_f = 0.822$, $k_f = 0.262$, $k_s = 0.0088$ with service age coefficient $k = 0.001$ are taken. When the concentration of slow reactant is increasing the free chlorine is marginally decreasing. When the concentration slow reactant is 10 free chlorine is reduced to zero in 17 hours while when the concentration slow reactant is 4.22 free chlorine become zero in 24 hours. Therefore, in this situation, the concentration of slow reacting agent decreases the ability of free chlorine marginally, the ability of free chlorine for clean the water as compared to the quantity of concentration of fast-reacting agents. Hence the higher concentration of slow reacting agents means that more chlorine is needed to purify the water distribution system. More dose of the chlorine is required to achieve and maintain the desirable level of chlorine residual.

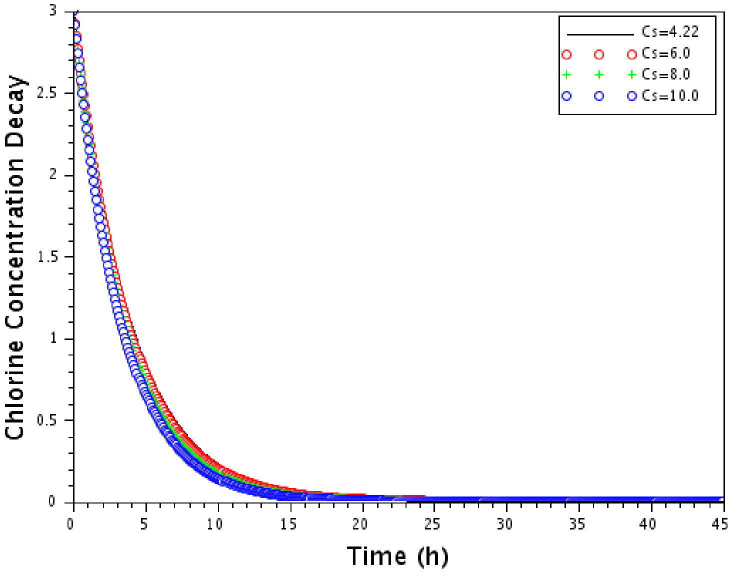

Figure 5. The Variation between chlorine decay rate with time for different values of slow reactant agents.

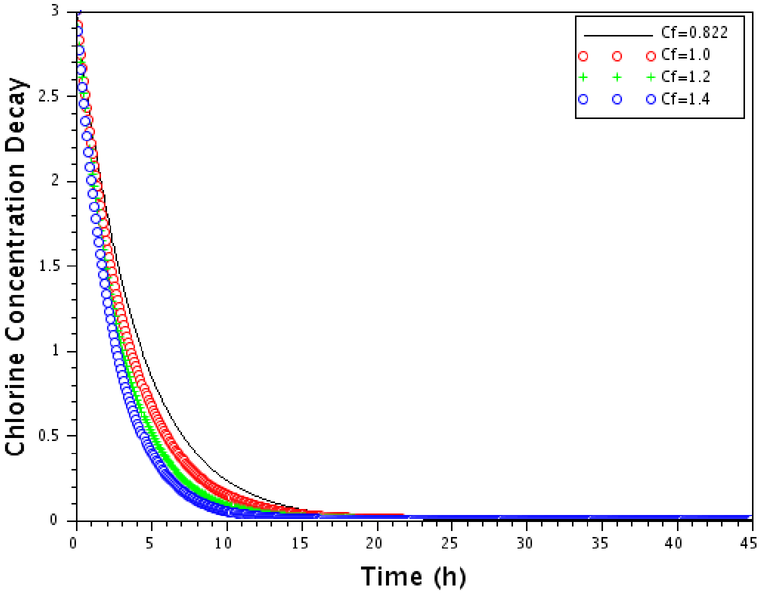

Figure 6. Variation between chlorine decay rate and time with different values of fast reactant agents.

Figure 6 is obtained for various values of concentration of fast-reacting agent C_f (0.822, 1.0, 1.2, 1.4), $k_f = 0.262$, k_s= 0.0088 and service age coefficient k = 0.001. When the value of the fast-reacting agent is higher chlorine concentration becomes zero at 15 hours. However, when $C_f = 0.822$, free chlorine becomes zero in 19 hours. Hence concentration of fast reacting agents increased as it consumed the chlorine rapidly. Hence more chlorine is required maintain the disinfection throughout out the network.

5. Experimental Observations

The sample was collected from Pithuwala, Dehradun, India water tank, 3 *mg/l* chlorine was used in the tank to check the chlorine residual in the water. The samples were also taken from old as well as newly built water tanks, and then collected data was compared with the data obtained by the mathematical model for the value of aging coefficient $k = 0.4$ and $k = 0.01$, the coefficient of correlation for 20 years old built water is computed with the value obtained from the mathematical model $k = 0.4$, it was found to be 0.978139, which shows a very strong positive correlation between the duration of the chlorine in the system and chlorine concentration decay. On the other hand, the correlation coefficient for the newly built water tank for the age coefficient $k = 0.01$ was found to be 0.9830450, which also shows a very strong positive correlation.

Thus, it is observed that in the newly built water tanks with relatively very small age coefficients ($k = 0.01$ in comparison to $k = 0.4$) nearly the same correlation coefficient as for the old water tank.

The regression model for 20-year-old water tank is represented by $C = -0.000715438\, \mathrm{t} + 2.865680473$, on the other hand for a newly built distribution system the regression model is given by $C = -0.000566783\, t + 2.992307692$, where C is the free chlorine in the system and t is the time duration in the systems.

The elasticity of the coefficients of parameters of regression models in both cases is negative and very weak but the constant terms are significantly positive. The computed regression models for 20 years old system and the newly built system reflecting the chlorine concentration on 20 locations along with the pipe are displayed in the following **Table 1**.

Furthermore, a comparison can be done with the help of **Figure 2** and **Table 1**.

Table 1. Chlorine concentration decay in old as well as newly built water tank.

Sample collecting location	*t* (Time per second)	Chlorine concentration in 20 years old age(mg/L)	Chlorine concentration in newly built water tank (mg/L)
S1	0	3	3
S2	200	2.9	3
S3	400	2.8	3
S3	600	2.6	2.9
S4	800	2.5	2.5
S5	1000	2.2	2.5
S6	1200	1.5	2
S7	1400	1.3	2
S8	1600	1.5	2
S9	1800	1.5	2
S10	2000	1.2	1.5
S11	2400	1.2	1.5
S12	2600	1.1	1.5
S13	2800	1.1	1.5
S14	3000	1	1.5
S15	3200	0.5	1
S16	3400	0.5	1
S17	3600	0.2	1
S18	3800	0.1	1
S19	4000	0	1
S20	4200	0	0.5

Figure 2 is compared with samples taken from the 20-year-old pipeline shown in **Table 1**. It is found that chlorine is reduced to 0.2 in 2 hours when k = 0.4 while chlorine is reduced to 0.2 in 1 hour, due to the concentration of fast reactant agent being less than the slow-reacting agents in the system. Furthermore, samples were also collected from the newly built pipelines and newly built tank, 3 mg /l chlorine was introduced in the tank and it was found that in 16 hours chlorine reduced to 0.5 while data obtained from a mathematical model, found that chlorine concentration became 0.5 in 5 hours, which is due to the concentration of slow reactant agent concentration present in the system or concentration of slow reacting agents reduces the speed of chlorine concentrations present in the systems.

6. Discussion

The mathematical model developed by Monteiro et al. (2015) provided the fast reaction rate and slow reaction rate, which they used without incorporating service age. while Bhadula et al. (2021) provided the service rate coefficient with chlorine concentration decay for an unsteady state mathematical model without using fast and slow reactant agents in their model. In this study, we used fast and slow-reactant agents along with the service age coefficient of the distribution system to investigate the chlorine concentration decay rate.

It was found that with the increase in the value of the service age coefficient, less free chlorine was available for cleaning the water due to a decrease in time for chlorine concentration decay.

With the fixed service rate coefficient and the change in the values of the slow reactant agent, it was found that with the increase in the slow reactant agent, the available free chlorine becomes zero in just 2 hours with the value of k_s= 0.6 in comparison to the value to free chlorine becoming zero in 7 hours with k_s = 0.1.

It was further found that with the fixed lower value of the service age coefficient (k = 0.001), it was noticed that the free chlorine was available for a longer duration to clean the water as compared to the fast reactant agent.

Very strong positive correlation was found between the duration of chlorine in the system and decay in the chlorine concentration.

7. Conclusion and Future Scope

The aging of infrastructure and biofilm can have an impact on the rate at which chlorine in water distribution systems degrades over time due to the reaction of chlorine with pollutants or chemical reaction with pipes and other system components. As they develop rough surfaces or corrosion. in the case of slow-acting surfaces. On the other hand, in the presence of slow-acting substances, like specific kinds of bacteria or inorganic chemicals, there is a relatively lower effect on the pace of chlorine degradation, in comparison to fast-reacting agents. Thus, free chlorine would be available to clean the water in the system as well as enhance the effectiveness of this infection over time. There seems to be almost direct proportionality between chlorine in the system and the decay in chlorine concentration. Future scope in the area of chlorine concentration decay in water pipeline networks will be depend on our understanding and ability to manage the water quality. This can be done by implemented such as machine learning and AI to predict chlorine decay. Furthermore, smart sensor can be used to provide the real time data and chlorine concentration levels along water pipeline. The material of the pipes used for water can be investigated to minimize chlorine decay as well as water loss in the system.

Conflicts of Interest

The authors declare that there are no conflicts of interest regarding the publication of this paper. The corresponding author confirms on behalf of all authors that there have been no involvements that might raise the question of bias in the work reported or in the conclusions, implications, or opinions stated.

AI Disclosure

The author(s) declare that no assistance is taken from generative AI to write this article.

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