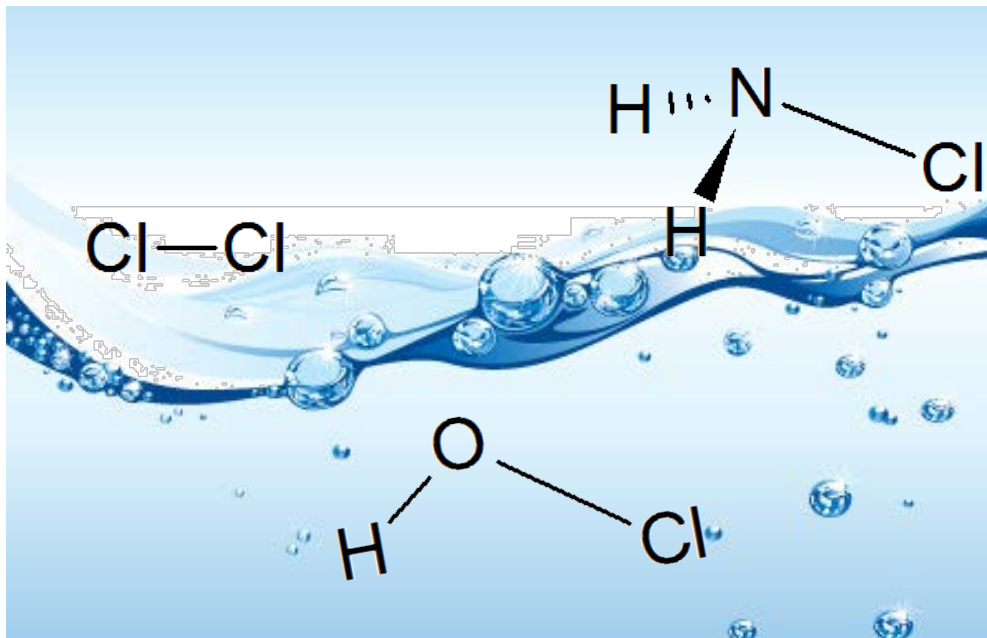


RECLAMATION

Managing Water in the West

Technical Memorandum No. MERL-2013-57

Effect of Chlorine vs. Chloramine Treatment Techniques on Materials Degradation in Reclamation Infrastructure



U.S. Department of the Interior
Bureau of Reclamation
Technical Service Center
Denver, Colorado

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The mission of the Bureau of Reclamation is to manage, develop, and protect water and related resources in an environmentally and economically sound manner in the interest of the American public.

BUREAU OF RECLAMATION
Technical Service Center, Denver, Colorado
Materials Engineering and Research Lab Group, 86-68180

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Treatment Techniques on Materials
Degradation in Reclamation
Infrastructure**

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Introduction

The Bureau of Reclamation, although historically focused on raw water transmission, is increasingly involved in water treatment and transport of treated water to local distribution systems. For the purpose of disinfection, this water is routinely chlorinated. Chlorine is a strong oxidant, and it has been shown that chlorination chemicals and their byproducts can adversely affect the performance and lifetime of infrastructure, specifically metallic pipeline materials, through corrosion. In addition, many water treatment systems have switched in the past decade from dosing with chlorine compounds to dosing with chloramines. This transition has been problematic in some distribution systems where chloramines have been shown to be more persistent and susceptible to causing corrosion.

A brief Reclamation review in 2012 compiled limited information for corrosion of copper, copper alloys, mild steel, and cast iron. Additionally, degradation of several elastomers by chlorine and chloramines was cited in the report. Recommendations were made for materials selection in mechanical components, piping, and valves. It was clear from the preliminary work that this would be a significant factor in materials selection when switching from untreated to treated water. It was unclear the degree to which chlorine versus chloramine could impact selection, and apparent that this issue warranted further investigations. This study expands upon the 2012 work and examines the current practices for chlorine and chloramine water treatment and state of knowledge of the effects of this treatment on corrosion.

Background

In the early 20th century, cities in the United States began treating drinking water with chlorine in order to combat water-borne diseases caused by pathogens such as bacteria, viruses, and protozoa [1]. This process has been highly successful and is responsible for largely eliminating many of these illnesses in the US. Treatment typically entailed dosing water with chlorine gas (Cl_2), liquid sodium hypochlorite solution (NaClO), or solid calcium hypochlorite ($\text{Ca}(\text{ClO})_2$). These compounds form hypochlorous acid (HOCl), or “free chlorine”, in water. Chlorine compounds are strong oxidizers, persist in water, and provide residual disinfection to prevent recontamination throughout the distribution system, making them useful in both primary and secondary disinfection processes.

Chlorine compounds are strongly reactive, a characteristic that makes them highly effective in rapid pathogenic disinfection. However, these compounds can also react with other natural organic compounds in water to form disinfection by-

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products (DBPs). Some of these DBPs are potentially harmful and, as such, have been regulated by the United States Environmental Protection Agency (USEPA). Although DBPs have not been proven to have adverse effect in humans at the levels at which they exist in drinking water, efforts have been made to limit human exposure through regulation. Particularly, two by-products of free-chlorine disinfection, trihalomethanes (THMs) and haloacetic acids (HAAs), have been determined to be carcinogenic with high exposure and have been identified for regulation under the Safe Drinking Water Act since 1979 and 1998, respectively, in the United States [2].

With the advent of DBP regulation, some water utilities transitioned from treatment with free-chlorine to treatment with chloramines. Monochloramine (NH_2Cl) offers several advantages over chlorine compounds, perhaps most importantly lower THM and HAA by-product concentrations. Monochloramine is also more persistent compared to free-chlorine compounds [3], allowing it to be more effective in maintaining disinfection throughout the distribution system. However, chloramine is a poor oxidant and not as effective in taste and odor control; it is, therefore, most often used as a secondary disinfectant.

As utilities transitioned from disinfection with free chlorine to chloramine, several investigations found increased levels of metal in the water system. The change in water chemistry with the transition from free chlorine to chloramine treatment, and the increased persistence of the chloramine and its byproducts, caused higher corrosion levels and increased leaching of the metal into the drinking water. In an extreme case, the city of Washington, D.C., transitioned from free chlorine to chloramine treatment in 2000. By 2001, elevated levels of lead were measured in tap water. Investigations by Edwards et. al. attributed the increase to interaction of the chloramine-treated water with lead pipes and lead solder [4-5]. Lead levels are regulated to an action level of 0.015 milligrams per liter (mg/L), and lead levels were observed over 85 times higher in tap water. Such elevated water lead levels caused a public health crisis, Congressional hearings, and class-action lawsuits citing adverse health and behavioral issues in children younger than six years of age at the time of contamination.

Although free-chlorine treatment is still the dominant disinfection process in the United States, chloramine treatment is prevalent as a secondary disinfectant, and it is practical to understand the pros and cons of each method. In addition, several secondary measures, such as pH control and phosphate addition, can be used to mitigate some of the unfavorable side effects for each treatment method. The following sections will outline each technique, its effect on corrosion in water infrastructure, and potential corrosion mitigation techniques.

Current Practices in Disinfection

Disinfection of drinking water in the United States is required by the Safe Drinking Water Act. National regulation of disinfection processes also apply to certain disinfection by-products, the leaching of lead and copper into water due to corrosion (Lead and Copper Rule [6]), and the transport, storage, and handling of certain chlorine-based disinfection chemicals. Individual states may choose to abide by federal regulations or to adopt more stringent standards. Individual utilities must develop a treatment program on a system-by-system basis taking into account local regulations, water quality, condition of the distribution infrastructure, and treatment capabilities. Characteristics of an “ideal” secondary disinfectant are listed in Table I [7].

Table I. Properties for an "Ideal" Disinfectant Residual [7]

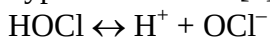
The “Ideal” Disinfectant Residual Provides:
• Protection against distribution system contamination
• An indication of distribution system upset
• Biofilm control
The “Ideal” Disinfectant Residual has the Following Chemical Characteristics:
• Easily measured on-site under field conditions
• Minimal to no interferences with common constituents in drinking water
• Generates minimal to no disinfection by-products
• Long-lasting
• Selectively reactive (minimal to no corrosion/reaction with dissolved metals, pipe materials, linings, etc.)
• Provides clear indication of contamination event (is chemically altered rather than consumed)
The “Ideal” Disinfectant Residual has the Following Operational/Physical Characteristics:
• Highly soluble in water
• Safely generated, transported, stored, and fed
• Cost-effective relative to the application (large- or small-scale)
The “Ideal” Disinfectant Residual has the Following Inactivation Capabilities:
• Effectively and efficiently inactivates wide range of organisms (bacteria, viruses, protozoa, algae, fungi)
• Effectively inactivates microorganisms present in the bulk water and those associated with particles/biofilm
• Achieves desired level of organism inactivation at doses that are safe for human consumption
The “Ideal” Disinfectant Residual has the Following Aesthetic Characteristics:
• Achieves desired level of organism inactivation without creating tastes and odors
• Overfeed can be detected by taste, odor, and/or color

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Free chlorine is the most widely used disinfectant in the United States for both primary and secondary disinfection. Chlorine compounds hydrolyze almost immediately in water by the following reactions [1]:

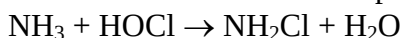


Hypochlorous acid will then dissociate, at $\text{pH} < 7.5$, to form hydrogen ions and hypochlorite ions [1]:



Because chlorine is such a strong oxidizer, it is rapidly utilized in many secondary reactions, such as bromide, iron, manganese, and sulfide oxidation, before disinfection can occur. After this initial demand has been met, free chlorine reaction rate slows and it persists in the water as a disinfectant residual. It is, therefore, necessary to dose chlorine compounds at a level high enough to achieve significant levels of free chlorine for disinfection. Utilities are recommended to measure both total chlorine concentration and free chlorine concentration when determining dosing levels for their treatment systems. Efficiency is dependent on disinfectant concentration, contact time, temperature and pH. The CT value, or concentration in parts per million (ppm) x contact time in minutes (min), is commonly used to gauge effectiveness. The lower the CT value, the more effective the disinfection agent.

Chloramines are not as reactive as chlorine, and, therefore, are not as effective as chlorines as primary oxidizers. They are often used as secondary disinfectants after primary treatment with chlorine followed by the controlled addition of ammonia to form monochloramine (NH_2Cl), often in the recommended chlorine to ammonia ratio of 4.5:1 to prevent nitrification.



The chloramine decay rate is much slower than that for chlorine, so, while higher concentrations are required, their persistence leads to longer contact time and effectiveness as a secondary treatment option. CT values for free chlorine and monochloramine effectiveness against *e. coli* and heterotrophic bacteria are compared in Table II [8]. The lower reactivity of chloramines can also be an advantage, as they are less likely to react with natural organic matter in the water, forming fewer DBPs and leading to fewer undesirable tastes and odors.

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Table II. Comparative efficiency of disinfectants for the production of 99% bacterial inactivation in oxidant demand-free systems [8]

Disinfectant	<i>Escherichia coli</i>			Heterotrophic bacteria		
	pH	Temp (°C)	CT mg/min l ⁻¹	pH	Temp (°C)	CT mg/min l ⁻¹
Hypochlorous acid	6.0	5	0.04	7.0	1–2	0.08 ± 0.02
Hypochlorite ion	10.0	5	0.92	8.5	1–2	3.3 ± 1.0
Chlorine dioxide	6.5	20	0.18	7.0	1–2	0.13 ± 0.02
	6.5	15	0.38	8.5	1–2	0.19 ± 0.06
	7.0	25	0.28			
Monochloramine	9.0	15	64	7.0	1–2	94.0 ± 7.0
				8.5	1–2	278 ± 46.0

Source: Adapted from LeChevallier, Cawthon & Lee (1988)

In 1996, amendment to the Safe Drinking Water Act required the USEPA to develop regulations to guide utilities in striking a balance between pathogen concentration and disinfection by-product concentration. The results were the Disinfectants and Disinfection Byproducts Rule (DBPR) and Enhanced Surface Water Treatment Rule (ESWTR). The maximum contaminant level (MCL) allowed by the Safe Drinking Water Act for THMs and HAAs are 0.080 and 0.060 milligrams per liter (mg/L), respectively. Both are regulated under combined standards that encompass several specific variations in the compound class. Stage 2 of the DBPR and the Long Term 2 ESWTR (LT2) took effect in 2012-2013. Stage 2 DBPR requires utilities to identify locations with the highest THM and HAA concentrations and use averaging of those locations for monitoring and calculation of the system compliance with the MCLs. Previously, MCL compliance was determined by the average of monitoring stations across the entire system. It is expected that more utilities will transition to chloramines for secondary treatment in order to comply with the new DBP regulations. Chlorine and chloramine concentrations are also regulated by the USEPA with maximum residual disinfection limits (MRDL) of 4.0 mg/L for each. Actual usage is typically much lower.

Effect of Disinfectants on Corrosion

As previously mentioned, chlorine has been the dominant disinfectant in the United States for many decades. With new technology and regulation, some utilities are choosing alternative disinfection methods, including incorporation of chloramines as secondary disinfection. With these process changes come changes in the water chemistry within the distribution system, changes that can upset the chemical balance and lead to problems such as corrosion. Free chlorine has a higher oxidation-reduction potential than chloramine. This strong oxidizing behavior produces a fairly stable scale in most metallic pipes that can be disrupted with the transition to chloramines. Chloramines are also known to cause nitrification if too much free ammonia is allowed to remain in the distribution

system [9-12]. The nitrification process has potential to locally lower the pH in alkaline waters and cause corrosion of metal pipeline and dissolution of cementitious pipeline liner materials. The following section will examine the effects of chlorine versus chloramine on corrosion and degradation in various pipeline materials.

Metals- Cu, Fe, Pb

Copper (Cu) and iron or steel (Fe) are common materials used in distribution piping. Lead (Pb) pipe for drinking water infrastructure has been banned in the United States since 1986, and went out of favor much before then, but can still be found in very old homes, or in Pb-based solder of copper pipe and in brass fittings. A 2002 survey sponsored by the Water Research Foundation determined that approximately 18.7 % of surveyed distribution system pipe material is unlined cast or ductile iron, 3.8 % is steel, and 3.0 % is “other” including copper. Customer service lines surveyed were 56.3 % copper, 3.3 % lead, 1.5 % steel, 1.2 % cast iron, and 2.3 % “other” including ductile and wrought iron and brass [13].

In 1991, the Lead and Copper Rule set lead and copper “action limits” at 0.015 and 1.3 (mg/L), respectively [6]. If more than 10% of tap water samples exceed the action level, water utilities must take additional steps to control the corrosivity of their water. The Lead and Copper Rule sets procedures for monitoring water distribution systems, as well as requirements that must be met should action limits be exceeded. These requirements include corrosion control treatment (CCT).

Iron content in drinking water has not been directly related to human health risks. The USEPA has issued a non-mandatory Secondary Maximum Contaminant Level (SMCL) of 0.3 mg/L as a guideline for water utilities. This is primarily to control undesirable aesthetic and taste effects of high iron concentrations in drinking water.



Figure 1. Corrosion research laboratory at the National Risk Management Research Laboratory [14]

It has long been known that the oxygen and disinfectants present in treated water can cause corrosion in metallic pipe [15-16]. The National Risk Management Research Laboratory (Figure 1) has a program to study corrosion, scaling, and metal mobility in drinking water distribution systems [14]. It is common in new homes and installations with metallic pipe for tap water to contain elevated concentrations of piping metals [17]. The high oxidation potential of disinfectants causes corrosion of these materials. Some of the metals dissolve into the water, but others are oxidized to form a scale on the interior of the pipe (Figure 2). As this scale grows over time, it acts as a protective layer and slows the rate of metal ions entering the water, thereby lowering and stabilizing the metallic ion concentration in tap water. Changes in water treatment techniques can change the water chemistry and cause disruption in the stability of the oxide scale. This can increase metal ion concentration, sometimes above MCLs, and require corrective action. There has been extensive investigation into the mechanisms of scale formation and disruption in various water chemistries, including the effect of transition from chlorine to chloramine disinfection.

Corrosion can be effected by many variables and the combinations thereof including pH, alkalinity, temperature, dissolved oxygen, natural organic matter, and the type of scale that is formed [11, 18-20]. For this reason, it is often difficult to directly compare corrosion mechanisms from studies done under varying conditions. However, there is general agreement on the mechanism for increased corrosion that some systems observe when switching from chlorine to chloramine. Chlorine is a very strong oxidizer and can form stable, passivating oxide scales, thus limiting release of metals into drinking water [4, 18-19, 21-22]. Chlorine-induced scale consists of iron (III) oxide (Fe_2O_3) in iron pipe, lead (IV) oxide (PbO_2) in lead pipe, and copper (II) oxide (CuO) in copper pipe. The corresponding scales that have been observed with chloramine, a weaker oxidizer, are composed of less stable compounds including iron (II) oxide (FeO) and hydroxide ($\text{Fe}(\text{OH})_2$) and carbonates in iron pipe, lead (II) carbonate (PbCO_3) in lead pipe, and copper (II) oxide (CuO) in copper pipe.

In the cases of iron and lead, the oxide scale developed during chloramine disinfection has a higher solubility in water than the chlorine scale, which could lead to a higher dissolution of the metal oxides into water. This solubility difference is especially significant when disinfection is changed from chlorine to chloramine: the passivating oxides can break down, leaching iron or lead into the water [19]. There is also suggestion that this effect can be reversed with transition from chloramines back to chlorines. The effect on oxide scale is not significant in copper pipe, and metal leaching is not as sensitive to the type of disinfectant. While some studies have observed a slight temporary increase in copper concentration of water, as the system stabilizes the copper leaching rate will also stabilize or even decrease [19, 23].



Figure 2. Scale formation on the interior of an iron pipe [14]

Polymeric Materials

The American Water Works Association published an extensive study in 2007 on the “Performance of Elastomeric Components in Contact with Potable Water” [24]. The study noted that elastomeric materials had been used for over 50 years in drinking water distribution systems for components such as valves, gaskets, seals, and fittings. Upon switching from treatment with free chlorine to treatment with chloramines, many utilities were observing premature failure of elastomeric components. The degradation appeared to be dependent not only on the disinfectant, but also on factors such as water pH, temperature, and the elastomer formulation. The AWWA study sought to establish quantitative methods for predicting life expectancy of elastomers in service, identify elastomer formulations with good performance in chloramine-treated water systems, and provide the water industry with guidelines for risk and finance management associated with elastomer performance. Researchers studied seven formulations in both laboratory and field tests: natural rubber, styrene butadiene rubber (SBR), nitrile (NBR), neoprene, ethylene propylene diene monomer, sulfur-cured (EPDM-S), and ethylene propylene diene monomer, peroxide-cured (EPDM-P). Researchers used a modified version of ASTM D6284-02 “Standard Test Method for Rubber Property—Effect of Aqueous Solutions with Available Chlorine and Chloramine” as the procedure for accelerated testing and ASTM D518-99 “Standard Test Method for Rubber Deterioration—Surface Cracking” to assess degradation [25-26].

Overall, the study found that EPDM-P performed “significantly better” than the other elastomers under exposure to chlorines and chloramines. However, all of the elastomers exhibited greater swelling, greater loss in hardness, and poorer tensile performance under exposure to chloramines than chlorine. An increase in disinfectant concentration caused an increase in degradation rate, and temperature effects were more pronounced at high concentrations, although these results varied in extremity between the various elastomer formulations. pH also played a

key role in elastomer performance, with degradation rates increasing at lower pH values where dichloramine is predominant over monochloramine.

Solvay Advanced Polymers conducted a test for chlorine resistance on three thermoplastic formulations: polysulfone, acetal copolymer, and 33% glass-filled nylon 6/6 [27]. The polysulfone showed little to no mass change under exposure to three concentrations of chlorine at 60°C, with the acetal copolymer exhibiting moderate loss, and, finally, the nylon 6/6 with severe degradation that would lead to total mass loss after 15 years exposure. While the study did not test chloramine exposure, nor the effects of pH or temperature, it does show that chlorine exposure must be taken into account when choosing a thermoplastic material for use in drinking water distribution systems.

Poly vinyl chloride (PVC) is generally considered to be nonreactive to oxidizing disinfectants, although very little information was found and no direct studies comparing PVC performance in chlorine versus chloramine. PVC is often used as a pipe material in water quality test system reactors.

Cementitious Materials

Cement surfaces are considered to be inert to degradation from chlorine and chloramine disinfectants. Cement-lined pipe is also often used in water quality test system reactors. However, there are some unsubstantiated suggestions on the internet that nitrification from excess ammonia in chloramine disinfection can cause formation of carbonic acid, locally lower pH, and cause dissolution of cements. Nitrification of various pipe materials, including new and old concrete, was studied in-depth in the doctoral thesis of Zhang [12]. The concrete was shown to leach calcium carbonate and affect the pH of the water samples. In water with an initial pH 7, nitrification was shown to cause further depression in pH, although this did not lead to an increase in concrete corrosion. At pH 8, nitrification had no effect on pH. This study seems to support the widely held belief that cementitious materials are largely non-reactive to oxidative disinfectants, even under nitrification conditions.

Corrosion Mitigation

Control of water pH and alkalinity are the primary methods that water utilities use to control corrosion in distribution systems. A 2000 study by Cantor et al. [15] showed that, while chlorine addition to a system caused an increase in corrosion in iron, lead, and copper pipe, the effect could be reversed in iron and copper by increasing the pH of the system to pH= 7.0-8.0. Lead corrosion could also be decreased in some circumstances.

Phosphate inhibitors have been shown to reduce dissolution of lead and iron [23, 28]. They are thought to form complexes on the metal pipe surface that are low solubility, although they may not necessarily decrease corrosion. Polyphosphates are commonly used to control iron, and orthophosphates are used to inhibit lead corrosion. pH must be carefully controlled to achieve optimum efficacy. The same study by Cantor [15] showed that addition of orthophosphate to chlorinated water decreased the corrosion in lead and iron pipes, but could cause an increase in copper corrosion at pH=8.0. Nguyen found that high levels of orthophosphate could control copper pitting, but low levels could actually worsen pitting corrosion [29]. Both polyphosphates and orthophosphate have been shown to reduce concrete corrosion at neutral pH [30].

Iron corrosion in water distribution systems has also been mitigated with silicate inhibitors. These form an insoluble coating on the pipe wall, thus preventing metal dissolution [13].

Conclusions

The effect of chlorine and chloramine use in secondary disinfection on corrosion in infrastructure materials is a complex process that is often system dependent. Chlorines have a higher oxidizing potential than chloramines, but residual chloramines are more persistent and may provide better disinfection from source to tap. Chlorines are known to form harmful disinfection by-products that, in some cases, may exceed maximum contaminant levels. Chloramines minimize DBP formation, but, due to their lower oxidation potential, may form higher solubility scales that can increase dissolution of metals. Elastomeric materials tend to be more strongly degraded by chloramines than chlorines. Corrosion issues, such as increased leaching of metals or degradation of polymers, may arise when transitioning between disinfection techniques due to their varying effects on water chemistry. Control of water pH and alkalinity and the addition of certain inhibitors can be effective in controlling corrosion in distribution systems.

Recommendations for Reclamation Infrastructure

As Reclamation projects arise that involve storage or transport of treated water, or direct treatment of water, it will be prudent to examine each on a case-by-case basis with corrosion considerations in mind. A close working relationship with local water utilities will be beneficial in defining design criteria. When connecting to existing systems, a thorough understanding of the water quality, disinfection process, and existing infrastructure materials will be needed in order

to design a compatible system. Similarly, if water utilities decide to transition to a new disinfection process, Reclamation will need to examine its affected infrastructure and ensure that care is taken in developing the new disinfection process so as not to introduce undue corrosion stress on the system. Materials selection for pipe and fittings during the design process will be the first step in mitigating corrosion issues. Mitigation measures such as pH control and phosphate addition may also be needed. Most importantly, a robust monitoring system compliant with USEPA regulations will be necessary to diagnose and avert potential problems before they lead to public health concerns or infrastructure failure.

Acknowledgements

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